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Distribution Coefficients for Radionuclides in Aquatic Environments

Dialysis Experiments in Marine Environments

Prepared by T. H. Sibley, A. E. Nevissi, W. R. Schell

College of Fisheries
University of Washington

Prepared for
U.S. Nuclear Regulatory
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Dialysis Experiments in Marine Environments

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Prepared by
T. H. Sibley, A. E. Nevissi, W. R. Schell

College of Fisheries
University of Washington
Seattle, WA 98195

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Distribution Coefficients for Radionuclides in Aquatic Environments. II. Studies in Marine and Freshwater Sediments Systems Including the Radionuclides ^{106}Ru, ^{137}Cs, and ^{241}Am. Annual Report: August 1977 - July 1978. W. R. Schell, T. H. Sibley, A. Nevissi, and A. Sanchez. NUREG/CR-0802. 70 pp.

Distribution Coefficients for Radionuclides in Aquatic Environments. III. Adsorption and Desorption Studies of ^{106}Ru, ^{137}Cs, ^{241}Am, ^{90}Sr, and ^{238}Pu in Marine and Freshwater Systems. Annual Report: August 1978 - July 1979. W. R. Schell, A. L. Sanchez, T. H. Sibley, and J. R. Clayton, Jr. NUREG/CR-0803. 76 pp.

Distribution Coefficients for Radionuclides in Aquatic Environments. Methodology. W. R. Schell, A. Sanchez, T. H. Sibley, A. E. Nevissi, J. R. Clayton, Jr., and E. A. Wurtz. NUREG/CR-1852. Vol. I. 27 pp.

ABSTRACT

The overall objective of this research program was to obtain new information that can be used to predict the fate of radionuclides that may enter the aquatic environment from nuclear power plants, waste storage facilities or fuel reprocessing plants. Important parameters for determining fate is the distribution of radionuclides between the soluble and particulate phases and the partitioning of radionuclides among various suspended particulates. This report presents the results of dialysis experiments that were used to study the distribution of radionuclides among suspended sediments, phytoplankton, organic detritus, and filtered seawater. Three experiments were conducted to investigate the adsorption kinetics and equilibrium distribution of ^{59}Fe, ^{60}Co, ^{65}Zn, ^{106}Ru, ^{137}Cs ^{207}Bi, ^{238}Pu, and ^{241}Am in marine system. Diffusion across the dialysis membranes depends upon the physico-chemical form of the radionuclides, proceeding quite rapidly for ionic species of ^{137}Cs and ^{60}Co but much more slowly for radionuclides which occur primarily as colloids and solid precipitates such as ^{59}Fe, ^{207}Bi, and ^{241}Am. All of the radionuclides adsorb to suspended particulates although the amount of adsorption depends upon the specific types and concentration of particulates in the system and the selected radionuclide. High affinity of some radionuclides, e.g., ^{106}Ru and ^{241}Am, for detritus and phytoplankton suggests that suspended organics may significantly effect the eventual fate of those radionuclides in marine ecosystems.

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DISTRIBUTION COEFFICIENTS FOR TRANSURANIC ELEMENTS IN AQUATIC ENVIRONMENTS

Dialysis Experiments in Marine Environments

INTRODUCTION

The hydrological transport and biogeochemical cycling of radionuclides in aquatic environments is dependent upon their distribution between the soluble and particulate phases. However, suspended particulates may behave quite differently depending upon particle size, density, or suitability as a food source for aquatic biota. Therefore, it is important to determine how radionuclide sorption differs for different types of particulates. Since it is difficult to sort particulates in natural assemblages, information on competitive sorption is generally obtained in laboratory experiments. We have investigated the uptake of radionuclides by suspended particulates in constant shaking experiments and in dialysis experiments (1-3). This report presents information on the competitive sorption of radionuclides by suspended sediments, phytoplankton and organic detritus in saltwater dialysis experiments.

The data are from three different experiments conducted under slightly different experimental conditions, with the radionuclides ^{59}Fe, ^{60}Co, ^{65}Zn, ^{106}Ru, ^{137}Cs, ^{238}Pu, and ^{241}Am. Briefly, the first experiment was designed to obtain the equilibrium distribution of radionuclides on sediment and phytoplankton; the second considered relatively long-term (11 day) uptake kinetics and the third concentrated on the initial uptake by phytoplankton, sediments and organic detritus. Each of these experiments is discussed separately below with a final discussion of the similarities and differences among the experiments. These experiments present information on the relative uptake of radionuclides under specific experimental conditions and may not apply directly to natural conditions. Nevertheless, the results should indicate which biogeochemical pathways are most significant for different radionuclides.

MATERIALS AND METHODS

The dialysis apparatus (Fig. 1) used for these experiments was adapted from similar equipment described by Barsdate (4) and Dawson and Duursma (5). In all our experiments, the outside compartment contained filtered (0.22 μm Millipore filters) water that was spiked with radionuclides and allowed to equilibrate for at least 10 days before the experiments began. It is important to attain equilibrium between stable and radioisotopes and among different physico-chemical species since the experiments are designed to show how the physico-chemical species found in natural environments may behave. After the equilibration period, dialysis sacs containing filtered, or distilled water were submerged in the radioactive solution and the suspended particulates were then added to the dialysis sacs. Initially only two dialysis sacs were used, one containing phytoplankton and the other containing suspended sediments (Fig. 1). In later experiments, two additional dialysis sacs were added in order to consider uptake of radionuclides by organic detritus and to have a control sac in order to study diffusion across the dialysis membrane. During the experiment the outer compartment is mixed with a magnetic stirrer and the contents of each dialysis sac are stirred and aerated with a glass stirring rod connected to a small electric motor.

Dialysis membranes are intended to exclude species above a specified molecular weight. For these experiments dialysis membranes with a 6000-8000 molecular weight cut-off were used. Thus, only ionic species, low molecular weight complexes, and small colloids can diffuse across the membrane. Samples were removed from the dialysis sacs and the outside chamber at predetermined times and filtered through Millipore or Nucleopore membrane filters to separate the soluble and particulate phases. The filters and a portion of the filtrate were then measured for radionuclides.

Samples for analyses were placed in 1.3 x 5 cm (2 dram) polyethylene vials. Gamma emitting radionuclides ^{59}Fe, ^{60}Co, ^{65}Zn, ^{106}Ru, ^{137}Cs, and ^{241}Am were measured with one of two systems. One system consists of a calibrated Ge(Li) detector with a computerized data reduction system. The other uses a Ge(Li) detector with a computer based multi-channel analyzer that integrates the counts for each gamma energy peak, subtracts background and prints out gross and net counts. The resolution (FWHM) of this Ge(Li) detector is 1.888 kev and the efficiency is 14% relative to 3" x 3" Na (Tl) detector.

After gamma-counting, the samples for ^{238}Pu analyses were spiked with ^{242}Pu and wet ashed with $HNO_3/HClO_4$. The plutonium isotopes were initially separated from the bulk of the matrix material by sorption from an 8 M HNO_3 solution on an ion exchange resin column (BIO-RAD-AGI-x8, 100-200 mesh). The plutonium solution was then purified, using the liquid ion exchange (triisooctylamine, TIOA) method by Butler.(6) The plutonium was electroplated onto a stainless steel disc and measured

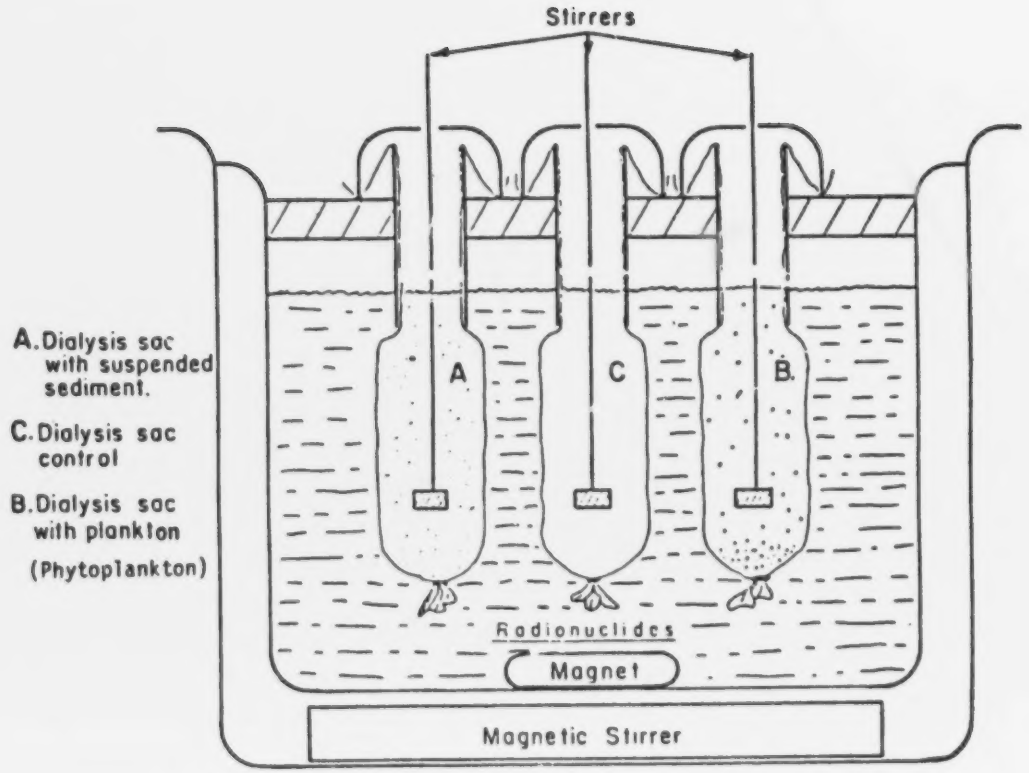

Figure 1. Schematic diagram of apparatus used for dialysis experiments.

with a silicon surface barrier alpha-detector and pulse-height analyzer.(7)

The phytoplankton species used in these experiments, Phaeodactylum tricornutum, was obtained from a culture maintained by Dr. Frieda Taub, College of Fisheries, University of Washington. Prior to use in the experiments, a stock algal suspension (8.6×10^6 cells/ml) was centrifuged at 6000 RPM for 15 min and resuspended in 32 °/oo NaCl solution. This process was repeated four times to remove the nutrient media and the final rinse was in 32 °/oo filtered seawater. The phytoplankton were then resuspended in filtered seawater before being added to the dialysis sac.

The suspended sediments were either natural sediments from Lake Nitinat, an anoxic fjord located on the western coast of Vancouver Island, British Columbia or the reference clay, calcium montmorillonite, obtained from the Georgia Kaolin Company. In each case, a concentrated stock solution was prepared with a known sediment concentration (mg/l) and a specified volume of the stock solution was added to the sediment dialysis sac.

Organic detritus was prepared from Zostera marina, a common marine macrophyte in Puget Sound. After washing and drying at 80°C, the Zostera was ground in a ball mill and sieved. For the experiments, a weighed amount of the <63 μm fraction was suspended in filtered seawater to obtain a stock suspension. A known volume of this suspension was then added to a dialysis sac so that the weight of detritus in the experiment could be calculated.

RESULTS AND DISCUSSION

The first experiment was designed to provide general information on the equilibrium distribution of ^{60}Co, ^{65}Zn, ^{137}Cs, ^{207}Bi, ^{238}Pu, and ^{241}Am on suspended sediments and phytoplankton in a seawater medium. These radionuclides were equilibrated with filtered seawater collected from a depth of 150 m in the Strait of Juan de Fuca, Washington. Membrane sacs filled with 50 ml of unspiked seawater from the same collection site were then suspended in the radioactive seawater. A phytoplankton sac was added with 5 ml of concentrated inoculum of Phaeodactylum tricornutum and the sediment sac contained 5 ml of filtrate (<0.45 μm) from the P. tricornutum culture and 31.3 mg of marine sediment from Lake Nitinat. A light source was provided to maintain the growth of the phytoplankton. The phytoplankton concentration at the end of the experiment was 7 x 10^6 cells/ml, and the total weight of the dry plankton was 6.8 mg. After 10 days samples were taken for analyses from each of the three compartments. Since Dawson and Duursma (5) demonstrated that equilibrium was established for most radionuclides within 100 hours, we assumed that ten days was sufficient to reach equilibrium in this experiment.

The concentrations of radionuclides in the soluble and particulate phase in each compartment are given in Table 1. Since the soluble concentrations are comparable in each compartment it is reasonable to assume that, except for ^{207}Bi, equilibrium was achieved in the 10-day period of the experiment. However, the soluble radionuclides, after passage through the membrane, were taken up by plankton and sorbed on the sediment particles. Therefore, the total concentration (soluble and particulate) may be different among the three compartments.

All the radionuclides, except ^{65}Zn and perhaps ^{137}Cs, are accumulated by phytoplankton. However, the concentration factors obtained for ^{65}Zn and ^{60}Co are two to four orders of magnitude lower than values reported by Lowman et al. (8). The concentrations of ^{238}Pu and ^{241}Am are particularly high in phytoplankton, relative to the dissolved concentration. It was reported previously that the major mode of plutonium and americium accumulation in P. tricornutum was a nonbiological process (9,10) and that adsorption, was the major mode of accumulation for both radionuclides. The radionuclides ^{207}Bi, ^{238}Pu, and ^{241}Am also accumulate on the suspended sediments, although the distribution coefficients for sediments are significantly lower than the concentration factors for phytoplankton (Table 2).

Table 1. Distribution of radionuclides after 10-day dialysis experiment.

Radionuclide	Spiked seawater compartment	
	Filtrate dpm/ml ± 2SD, %	Particulate dpm/ml ± 2SD, %
^{241}Am	25 ± 14.2	25 ± 1.8
^{207}Bi	*	0.78 ± 17.3
^{137}Cs	827 ± 1.1	208 ± 5.3
^{65}Zn	1533 ± 1.3	27 ± 4.6
^{60}Co	1108 ± 1.1	21 ± 3.6
^{238}Pu	0.30 ± 0.02	*

	Phytoplankton sac compartment	
	Filtrate dpm/ml ± 2SD, %	Phytoplankton on filter dpm/g ± 2SD, %
^{241}Am	29 ± 5.6	2.77×10^5 ± 2.2
^{207}Bi	2 ± 50	299 ± 18
^{137}Cs	701 ± 0.7	*
^{65}Zn	1669 ± 0.8	465 ± 0.6
^{60}Co	1107 ± 0.7	1.73×10^4 ± 1.4
^{238}Pu	0.25 ± 0.02	488 ± 22.2

	Sediment sac compartment	
	Filtrate	Sediment on filter
^{241}Am	27 ± 5.2	2615 ± 2.0
^{207}Bi	*	41 ± 24.4
^{137}Cs	836 ± 0.6	75 ± 19.3
^{65}Zn	1511 ± 0.7	566 ± 11.7
^{60}Co	1095 ± 0.6	893 ± 6.0
^{238}Pu	0.32 ± .03	20.2 ± 5.0

*Below minimum detection.

Table 2. Concentration factors for phytoplankton and distribution coefficients for sediments as determined in a dialysis experiment with Eastern Pacific seawater.

Radionuclide	Concentration factor for phytoplankton dpm/g phytoplankton / dpm/ml filtrate	Distribution Coefficient for sediment dpm/g sediment / dpm/ml filtrate
^{241}Am	9447	98
^{207}Bi	128	*
^{137}Cs	*	0.1
^{65}Zn	0.3	0.4
^{60}Co	16	0.8
^{238}Pu	1952	63

*Counting data for particulate or soluble fraction is below minimum detectable limit.

A second experiment was conducted to determine the rate of transfer of radionuclides across the dialysis membranes and of sorption to phytoplankton and suspended sediments. The dialysis experimental setup was similar to that reported previously except that the size of the apparatus was scaled up to accommodate repetitive sampling and a control dialysis sac free of plankton and sediment was added. Approximately 3,000 ml of spiked seawater was placed in the outer compartment and three dialysis sacs with 100 ml of distilled water were submerged in the seawater. Five ml of a Phaeodactylum tricornutum culture (2×10^5 cells/ml) was added to the phytoplankton sac and 5 ml of filtrate from the algal culture was added to the control and sediment sacs; the sediment sac contained 765 mg of sediment from Lake Nitinat. The experiment was conducted at pH = 7.5 and the equilibrium salinity was 29.1 $^{0}/_{00}$. Samples were collected from all four compartments at the beginning of the experiment and after 1, 2, 4, and 11 days and analyzed for radioactivity in the soluble (<0.22 µm) and particulate phase.

The experimental results are shown in Figs. 2-6 in terms of radionuclide concentration in the soluble and particulate phases of each compartment as a function of time. Figure 2 shows the changes in concentration for ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi, and ^{241}Am in the outer, spiked seawater compartment. Changes in the concentration of ^{238}Pu are shown in Fig. 3. During the first two days there is a rapid decline in the concentration of all radionuclides indicating that the radionuclides are diffusing into the unspiked chambers. Except for ^{207}Bi, the decrease in concentration in the outer compartment results primarily from soluble species diffusing across the dialysis membranes and into the dialysis

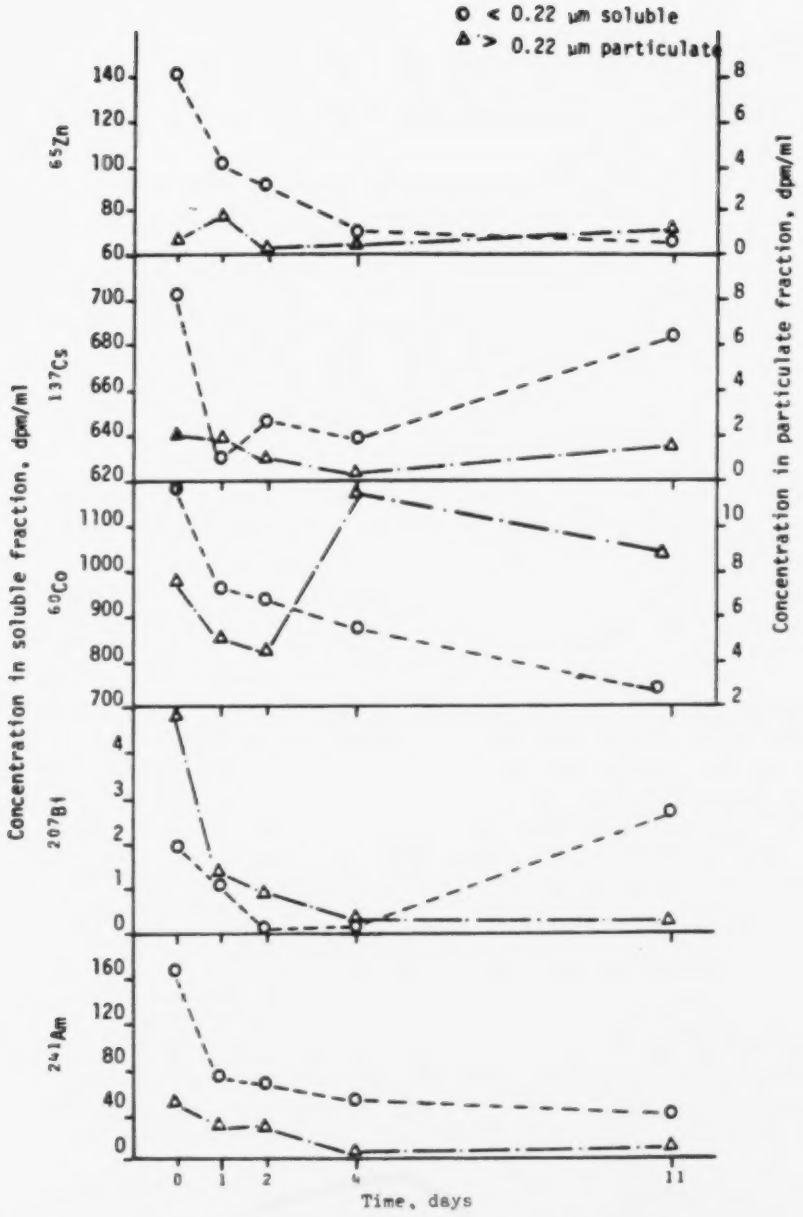

Figure 2. Concentration of radionuclides in soluble and particulate phases outside the dialysis sacs on days 0, 1, 2, 4, and 11 of a dialysis experiment.

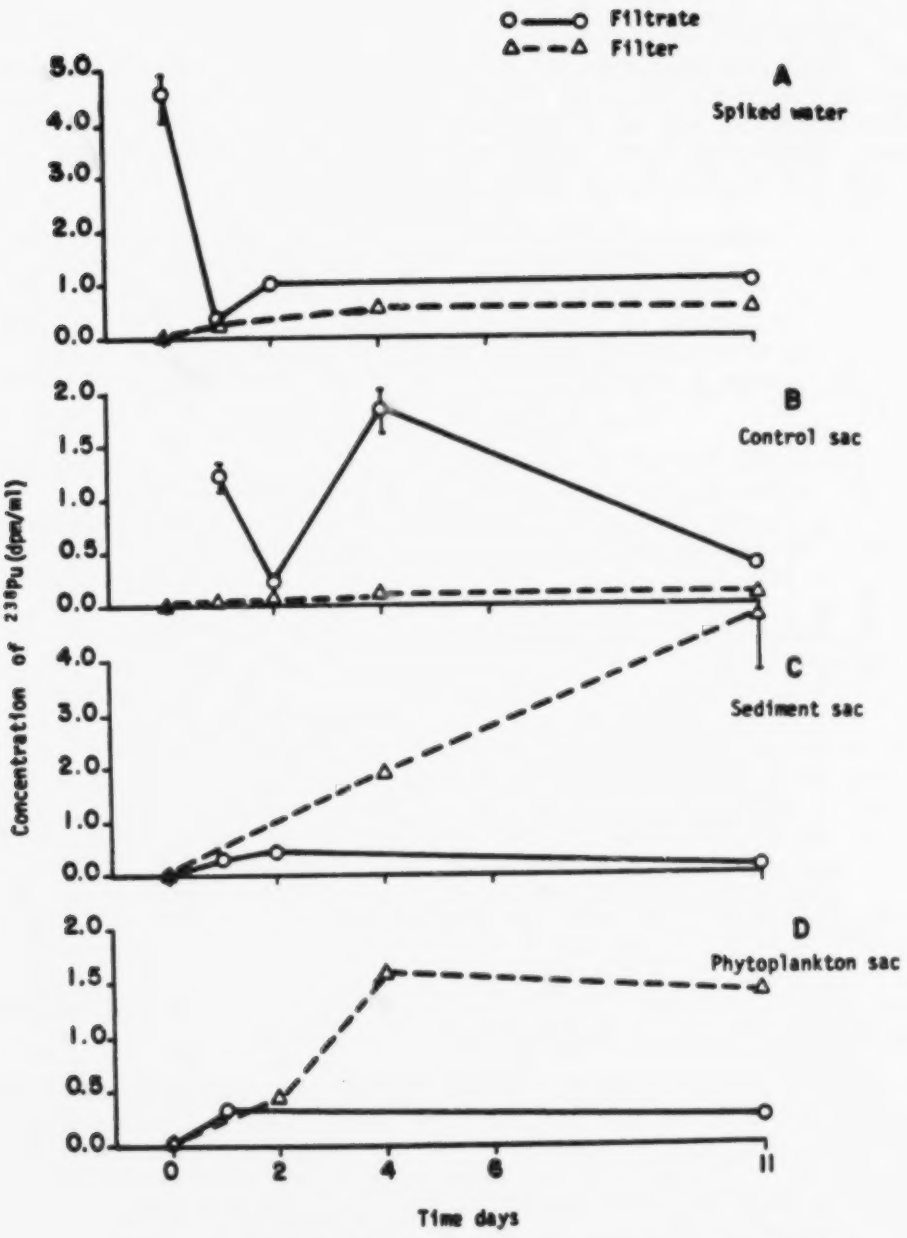

Figure 3. Concentration of ^{238}Pu in the soluble and particulate phases on days 0,1,2,4, and 11 of a dialysis experiment.

sacs. Some loss may also result from adsorption of soluble or particulate radionuclides onto the beaker or the dialysis membranes. After the fourth day there are no significant changes, i.e., within counting errors, in the concentration of any of the radionuclides in either the soluble or the particulate phase.

It is important to note the scale used for the particulate and soluble data for ^{65}Zn, ^{137}Cs, and ^{60}Co. For these three radionuclides the radionuclide concentration in the particulate phase is generally less than 1% of that in the soluble phase. Therefore, the apparent increase in concentration of particulate ^{60}Co between day 2 and 4 (Fig. 2) is only a negligible fraction of the total ^{60}Co concentration present.

Inside the control dialysis sac (Fig. 4) there was a significant increase in the soluble concentration of all radionuclides, except ^{207}Bi, during the first day due to the transport of soluble radionuclides. The concentration of ^{241}Am continued to increase slightly until day four. However, the concentration of soluble ^{241}Am declines between days 4 and 11 and the differences between days 2 and 11 did not exceed the propogated counting errors. For ^{65}Zn, ^{60}Co, and ^{137}Cs the soluble concentrations were unchanged or declined slightly from day 1 until the end of the experiment. There is no apparent reason for the high concentration of ^{238}Pu in the control sac on day 4 (Fig. 3). Similarly to ^{241}Am there was a decline in soluble ^{238}Pu concentration after day 4 and the concentration on day 11 is not significantly different than the concentration after the first two days. There was no detectable concentration of soluble ^{207}Bi in the control dialysis sac (Fig. 4) until the fourth day. However, the appearance of particulate ^{207}Bi inside the control sac indicates that soluble ^{207}Bi from the outer chamber diffused across the dialysis membrane and then precipitated. The decrease in the concentration of particulate ^{207}Bi in the outer chamber also suggests this mechanism. It appears that the diffusion of soluble ^{207}Bi from the outer chamber releases ^{207}Bi from the particulate phase and produces a decrease in the concentration of particulate ^{207}Bi.

Both ^{241}Am and ^{207}Bi inside the control sac are found in the particulate phase after one day and reach an apparent equilibrium between the particulate and soluble species by day 4; however, the concentrations of particulate ^{137}Cs and ^{60}Co increased significantly between days 4 and 11 indicating that these radionuclides did not reach equilibrium during the first four days. As with the outside compartment, in the control sac only a small fraction (<5%) of the total concentration of ^{137}Cs and ^{60}Co is in the particulate phase. It seems likely that the particulate ^{137}Cs and ^{60}Co are in fact adsorbed on radiocolloids or metal oxides that are precipitating within the dialysis sac. Since precipitation will not occur until the solubility product of a solid species is exceeded and the elements must diffuse into the dialysis sac before precipitating, the increase in particulate ^{137}Cs and ^{60}Co after 4 days probably results

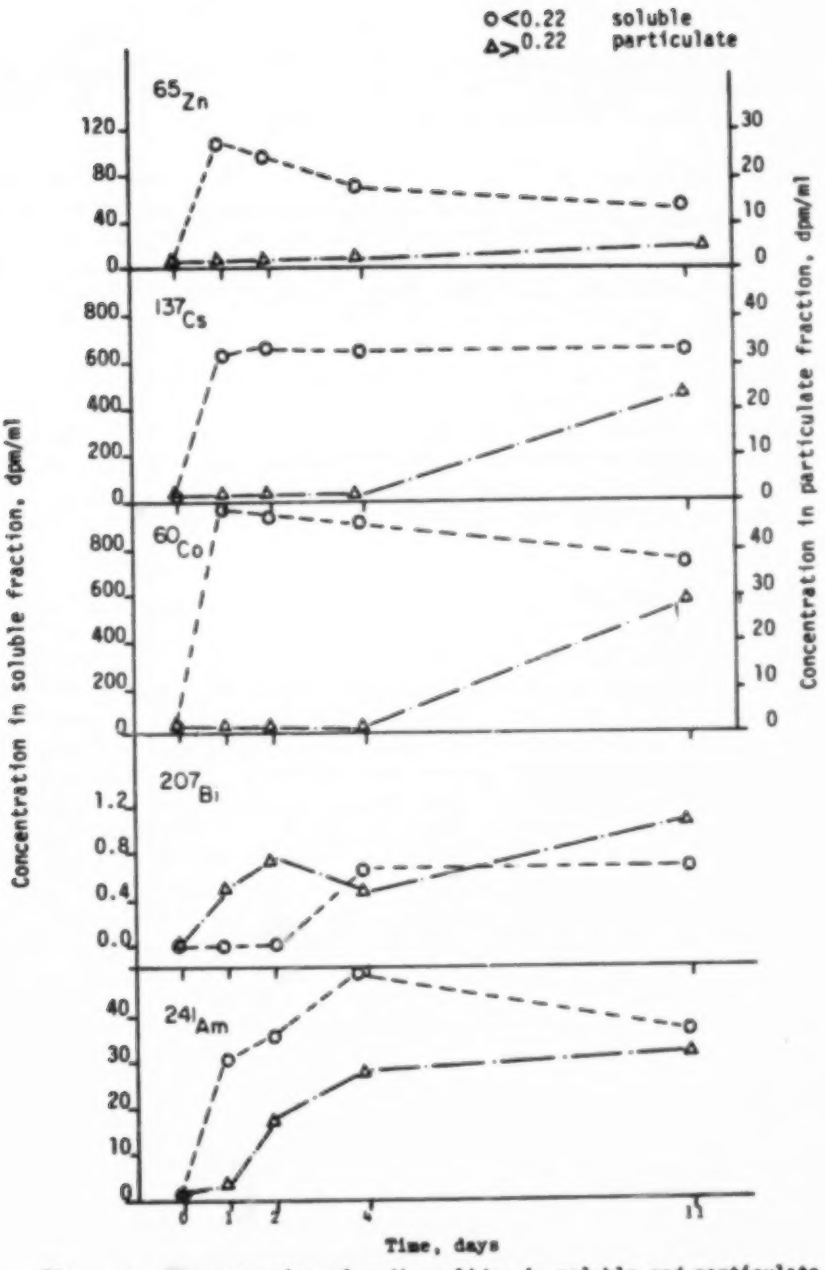

Figure 4 . Concentration of radionuclides in soluble and particulate phases within the control dialysis sac on day 0, 1, 2, and 11 of a dialysis experiment.

from the slow kinetics of precipitation. The concentrations of particulate ^{65}Zn and particulate ^{238}Pu in the control dialysis sac are low and relatively constant throughout the experiment.

The distribution of ^{60}Co, ^{65}Zn, ^{137}Cs, ^{207}Bi, and ^{241}Am between soluble and particulate phases in the plankton and sediment sacs is shown in Figs. 5 and 6; similar information for ^{238}Pu as shown in Fig. 3. Sediments accumulated significantly higher concentrations of all these radionuclides than the phytoplankton did. However, except for ^{60}Co and ^{137}Cs, greater than 80% of the total activity is found in the particulate phase in both the sediment and phytoplankton chambers. Less than 1% of the ^{60}Co and ^{137}Cs is taken up by the phytoplankton during the first four days although approximately 2% of ^{137}Cs and 13% of ^{60}Co occur as particulates after 11 days. This increase corresponds to the increase noted in the control dialysis sac between days 4 and 11 and may represent sorption of colloids rather than biological uptake of ^{60}Co and ^{137}Cs. In the sediment sac 35%-45% of ^{137}Cs and greater than 90% of ^{60}Co is particulate.

Soluble species of radionuclides appear to reach equilibrium among the different chambers of the dialysis apparatus within four days. In fact, soluble ^{137}Cs is in equilibrium after 1 day. There are, however, significant increases in the particulate concentration of most radionuclides between day 4 and 11. This may result from slow ion substitution reactions at solid surfaces or, as mentioned previously, from the precipitation of solid metal oxides where the radionuclides can sorb. In the sediment sac all the radionuclides showed significant increases in particulate concentration between days 4 and 11. A similar increase was observed for all radionuclides except ^{238}Pu in the phytoplankton chamber. This indicates that as radionuclides adsorb onto the particulates a net flux of radionuclides into the dialysis sac continues. It may also indicate that the binding capacity of the particulates is not saturated in 11 days.

The distribution coefficients for sediments and concentration factors for phytoplankton were not calculated because of poor counting statistics, and consequently large propagated errors, for radionuclides in the water phase. However, the specific activity (dpm/g) was calculated. The concentrations of ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi, and ^{241}Am per unit weight of sediment and phytoplankton are shown in Fig. 7; specific activity for ^{238}Pu is shown in Fig. 8. The concentration of ^{65}Zn and ^{238}Pu on phytoplankton is relatively constant throughout the experiment. There is, however, some variation in the specific activity of ^{60}Co, ^{207}Bi, and ^{241}Am on phytoplankton. No values are reported for the concentration of ^{137}Cs on phytoplankton prior to day 11 since the concentrations were below the analytical detection limits. The concentrations of ^{241}Am, ^{207}Bi, and ^{60}Co are approximately equivalent for both sediments and phytoplankton. However, significantly greater quantities of ^{137}Cs are accumulated by sediments, while phytoplankton have a slightly higher concentration of ^{65}Zn and ^{238}Pu.

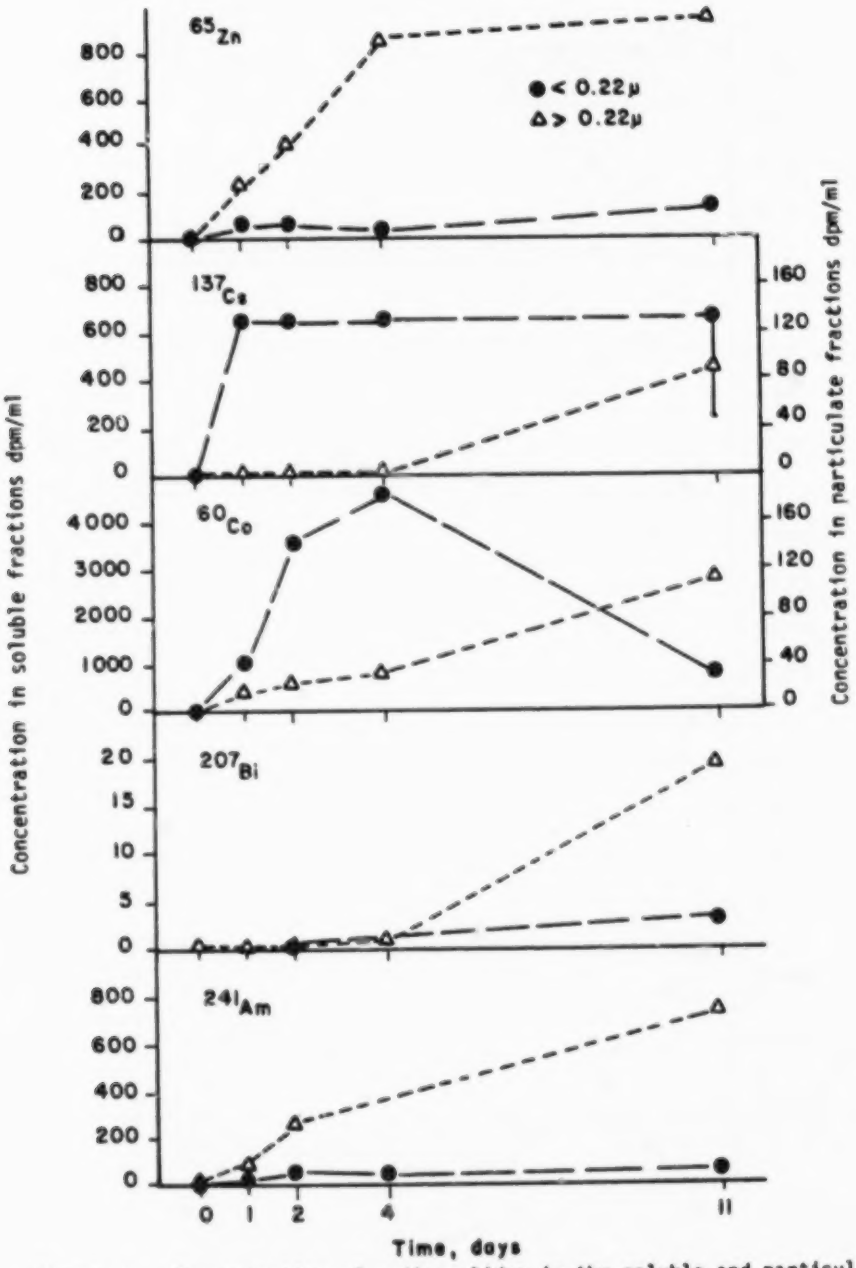

Figure 5. Concentration of radionuclides in the soluble and particulate phases within the plankton dialysis sac on days 0, 1, 2, 4 and 11 of a dialysis experiment.

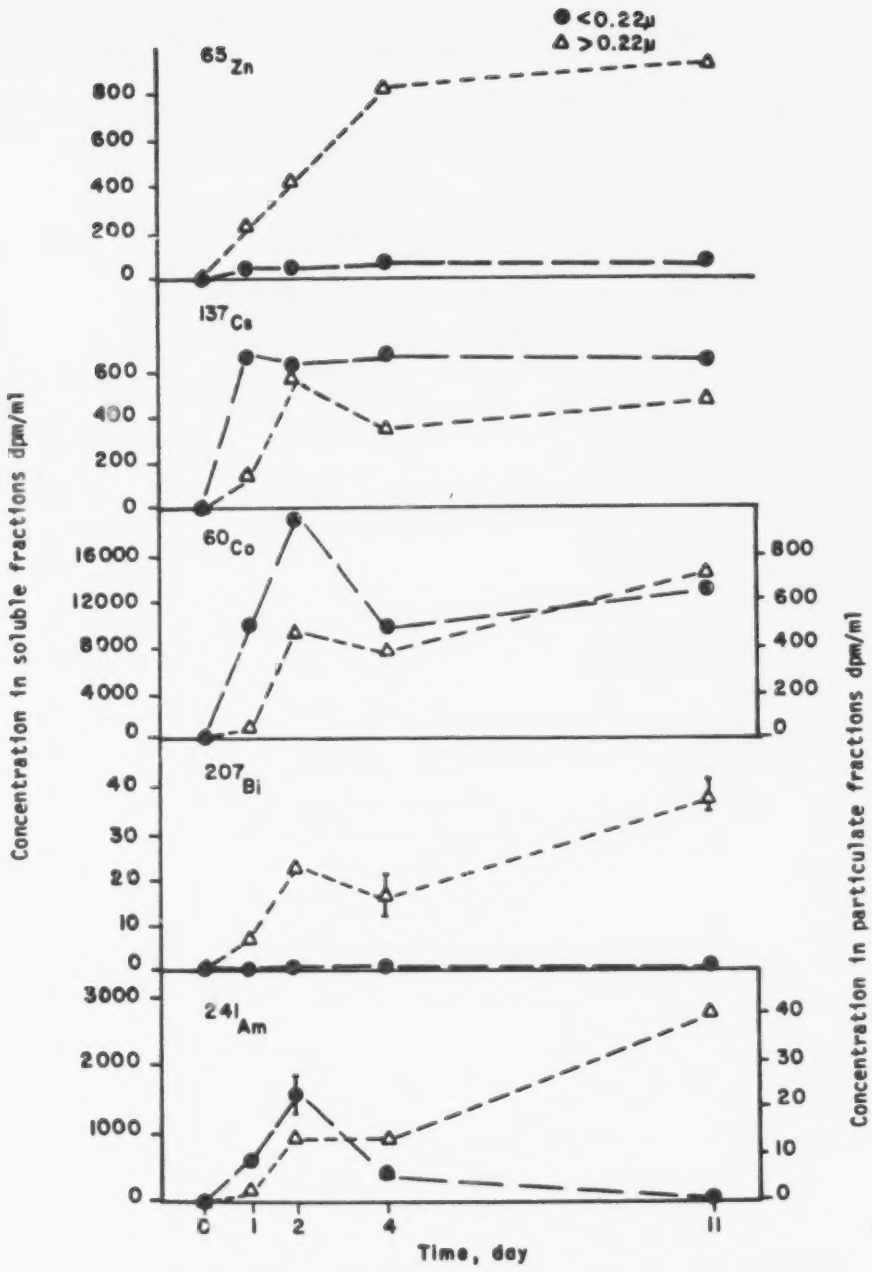

Figure 6 . Concentration of radionuclides in the soluble and particulate phases within the sediment sac on days 0, 1, 2, 4 and 11 of a dialysis experiment.

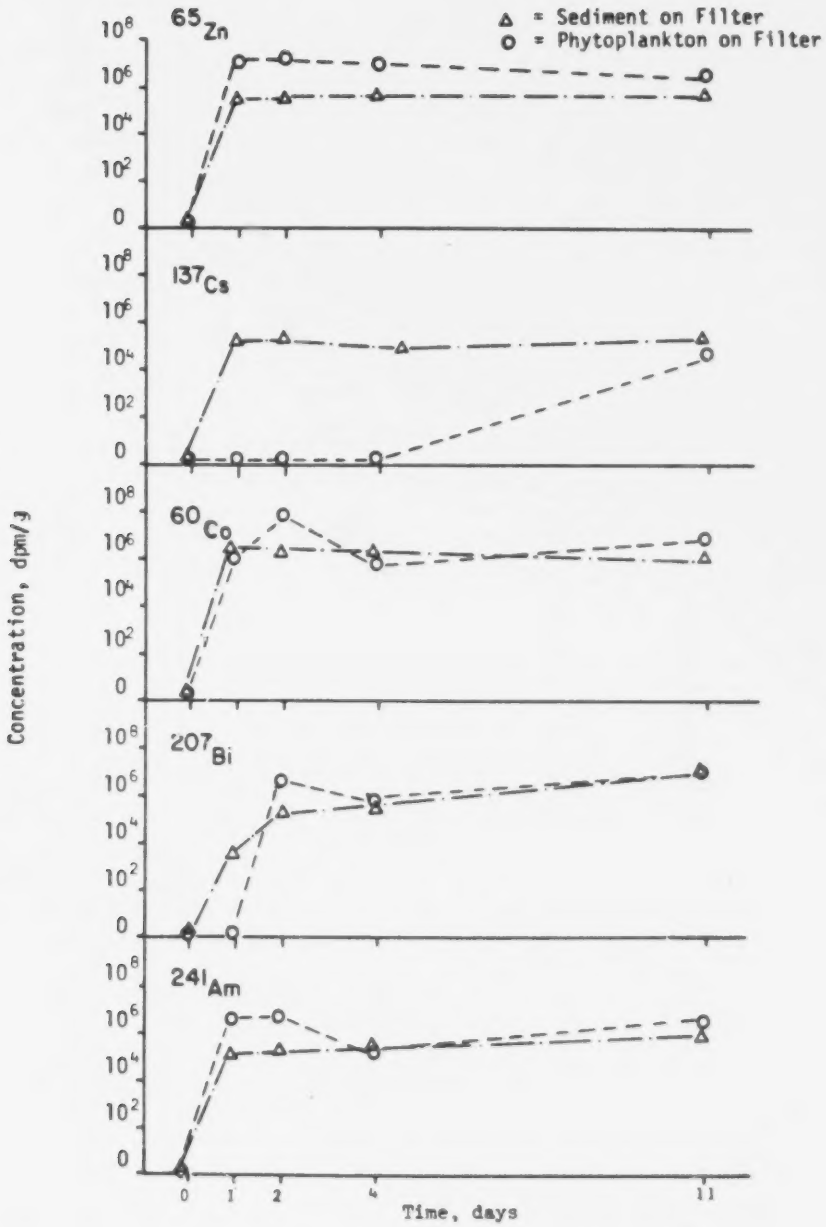

Figure 7 . Concentration of ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi and ^{241}Am per unit weight of sediment and plankton on days 0, 1, 2, 4 and 11 of a dialysis experiment.

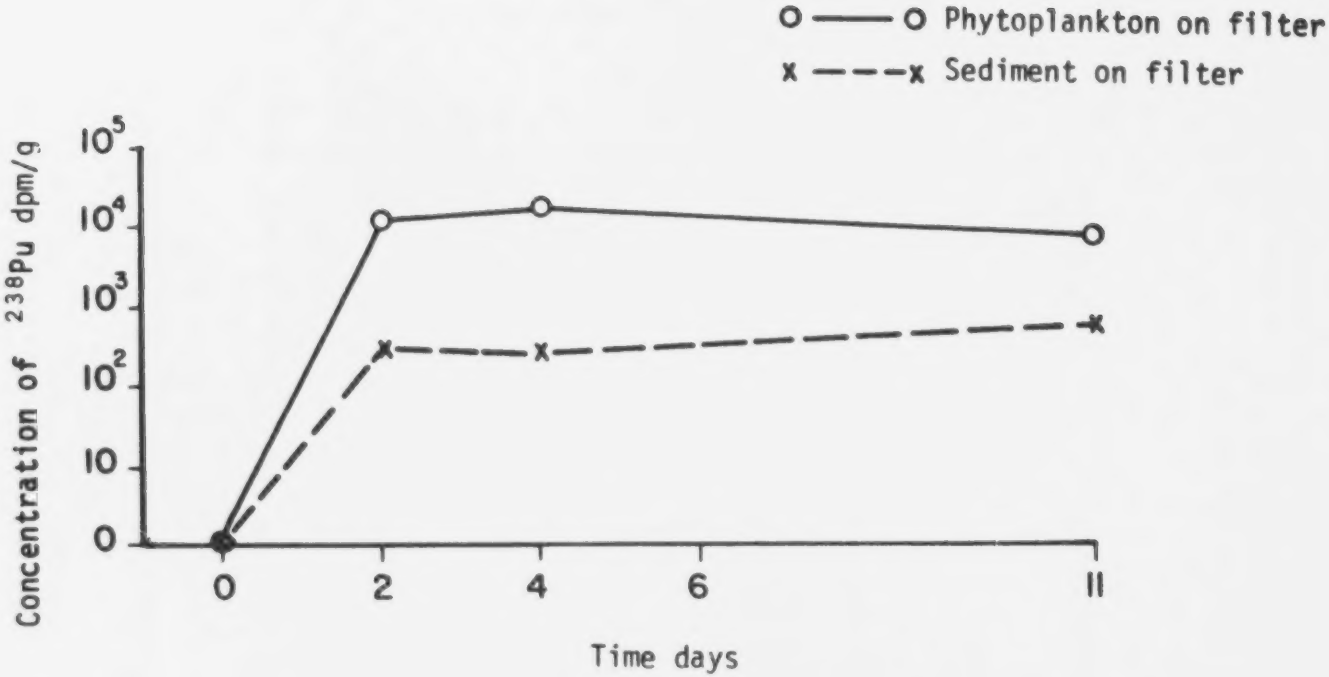

Figure 8. Concentration of ^{238}Pu per unit weight of sediment and plankton on days 0, 1, 2, 4 and 11 of a dialysis experiment.

The specific activity associated with phytoplankton and sediment in this experiment are much higher than in the previous experiment (Table 1, Fig. 7, 8). Several different experimental conditions including salinity, pH, temperature and the concentration of phytoplankton and sediment may contribute to the observed differences in specific activity. The temperature and salinity differences between the two experiments should produce insignificant, if any, differences. Some increase in specific activity of phytoplankton may have occurred due to the lower phytoplankton density (0.9×10^4 cells/ml) in the second experiment compared to the first (7×10^6 cells/ml). However, the sediment concentration is a factor of 6 higher in the second experiment. Therefore, differences in sediment concentration cannot explain the higher specific activities found in the second experiment. The most likely explanation for the differences in specific activity is different pH values during the two experiments. At the end of the second experiment the pH was approximately 7.5. Although pH was not measured following the first experiment a value significantly lower than 7.5 would explain the different results obtained in these two experiments. Such a difference in pH would also explain why the concentration factors for phytoplankton in the first experiment are so low compared to Lowman et al. (8).

Mass balance calculations were made for the radionuclides in this experiment at day 11. These calculations include the activity in the particulate and soluble phase for each of the compartments at day 11, the activity removed by sampling during the experiment and the activity adsorbed to miscellaneous apparatus. The latter includes activity adsorbed to the dialysis membranes and that removed from the glassware by three washings with hot HNO_3. Results of these calculations are provided in Tables 3-8. Approximately 100% of ^{60}Co, ^{65}Zn, ^{137}Cs, and ^{207}Bi was recovered. However, only 68% of ^{241}Am and 21% of ^{238}Pu was recovered. These radionuclides adsorb very strongly to glassware and probably were not removed by the HNO_3 washes.

In terms of the percent recovered, the major portion of ^{207}Bi, ^{60}Co, ^{137}Cs, ^{238}Pu, and ^{65}Zn occurs in the soluble fraction of the outer compartment. However, ^{241}Am has ≈ 47% of the recovered radionuclide in the particulate phase of the sediment sac and another 12% in the particulate phase of the plankton sac. To better compare the overall distribution of radionuclides in this experiment, Table 9 presents the percent of the total recovered activity in each compartment for each rationuclide.

Table 3. Mass balance calculations for ^{241}Am after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	1.14×10^5	1.10×10^4	26.1	2.6
	filter	3.60×10^4	2.16×10^3	8.2	0.5
b, inside control sac	filtrate	2.84×10^3	2.88×10^2	0.6	0.1
	filter	2.31×10^3	1.28×10^2	0.5	0.0
(2) Dialysis sac contents					
a, sediment sac	filtrate	0.00	0.00	0.0	0.0
	filter	2.06×10^5	1.14×10^3	47.1	1.3
b, plankton sac	filtrate	3.49×10^3	3.70×10^2	0.8	0.1
	filter	5.4×10^4	6.52×10^2	12.3	0.4
(3) Subsamples		1.95×10^4	1.47×10^2	4.4	0.1
(4) Flask washdown and dialysis sacs		2.18×10^3	4.71	0.5	0.0
Total of (1), (2), (3), and (4)		4.37×10^5	1.13×10^4	100.7	3.0
Amount added+		6.45×10^5	2.13×10^4		
% Accounted for				67.8	2.8
% Unaccounted for				-32.2	2.8

*Single sample error values are one-sigma, propagated, counting error.

+Amount added = spiked water at day 0.

Table 4. Mass balance calculations for ^{137}Cs after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	2.00×10^6	5.21×10^4	89.3	3.1
	filter	0.00	0.00	0.0	0.0
b, inside control sac	filtrate	4.97×10^4	1.32×10^3	2.2	0.1
	filter	1.71×10^3	1.85×10^2	0.1	0.0
(2) Dialysis sac contents					
a, sediment sac	filtrate	4.73×10^4	1.11×10^3	2.1	0.1
	filter	3.48×10^4	1.30×10^3	1.6	0.1
b, plankton sac	filtrate	4.95×10^4	1.16×10^3	2.2	0.1
	filter	7.17×10^3	3.03×10^3	0.3	0.1
(3) Subsamples		4.64×10^4	3.12×10^2	0.7	0.0
(4) Flask washdown and dialysis sacs		1.02×10^1	1.40	0.0	0.0
Total of (1), (2), (3), and (4)		2.24×10^6	5.23×10^4	98.5	3.1
Amount added+		2.12×10^6	4.77×10^4		
% Accounted for				105.7	3.4
% Unaccounted for				+ 5.7	3.4

*Single sample error values are one-sigma, propagated, counting error.

+Amount added = spiked water at day 0.

Table 5. Mass balance calculations for ^{238}Pu after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	3.17×10^3	2.85×10^2	77.7	9.2
	filter	1.22×10^2	3.27×10	3.0	0.8
b, inside control sac	filtrate	2.09×10	3.08×10	0.5	0.1
	filter	3.31	3.10×10^{-1}	0.1	0.4
(2) Dialysis sac contents					
a, sediment sac	filtrate	7.01	1.93	0.2	0.0
	filter	3.76×10^2	9.63×10	9.2	2.5
b, plankton sac	filtrate	1.59×10	3.08	0.4	0.1
	filter	1.08×10^2	7.37×10	2.6	1.8
(3) Subsamples		1.00×10^2	2.73	2.5	0.2
(4) Flask washdown and dialysis sacs		1.57×10^2	3.25×10	3.9	0.3
Total of (1), (2), (3), and (4)		4.08×10^3	3.13×10^2	100.0	9.7
Amount added†		1.93×10^4	2.46×10^3		
% Accounted for				21.1	3.1
% Unaccounted for				-78.9	3.1

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table 6. Mass balance calculations for ^{207}Bi after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	7.73×10^3	3.26×10^3	64.4	15.7
	filter	0.00	0.00	0.0	0.0
b, inside control sac	filtrate	0.00	0.00	0.0	0.0
	filter	7.70×10^1	2.28×10^1	0.6	0.2
(2) Dialysis sac contents					
a, sediment sac	filtrate	0.00	0.00	0.0	0.0
	filter	2.85×10^3	2.75×10^2	22.3	6.1
b, plankton sac	filtrate	2.46×10^2	8.55×10^1	1.9	0.8
	filter	1.47×10^3	1.21×10^2	11.5	3.1
(3) Subsamples		3.62×10^2	2.81×10^1	2.8	0.3
(4) Flask washdown and dialysis sacs		7.40×10^1	9.7×10^{-1}	0.6	0.2
Total of (1), (2), (3), and (4)		1.28×10^4	3.27×10^3	104.1	17.1
Amount added†		1.47×10^4	6.24×10^2		
% Accounted for				87.1	22.6
% Unaccounted for				-12.9	22.6

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table 7. Mass balance calculations for ^{65}Zn after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside	filtrate	1.92×10^5	7.78×10^3	44.7	2.0
dialysis sacs	filter	3.27×10^3	6.80×10^2	0.8	0.2
b, inside	filtrate	4.10×10^3	1.90×10^2	0.9	0.1
control sac	filter	3.62×10^2	4.11×10^1	0.1	0.0
(2) Dialysis sac contents					
a, sediment	filtrate	4.84×10^3	1.67×10^2	1.1	0.0
sac	filter	1.11×10^5	6.69×10^2	25.8	0.5
b, plankton	filtrate	1.04×10^4	2.44×10^2	2.4	0.1
sac	filter	7.28×10^4	4.37×10^2	16.9	0.3
(3) Subsamples		3.15×10^4	1.01×10^2	7.3	0.1
(4) Flask washdown and dialysis sacs		1.05×10^2	1.27	0.0	0.0
Total of (1), (2), (3), and (4)		4.30×10^5	7.85×10^3	100.0	2.1
Amount added+		4.27×10^5	9.39×10^3		
% Accounted for				100.7	2.9
% Unaccounted for				+0.7	2.9

*Single sample error values are one-sigma, propagated, counting error.

+Amount added = spiked water at day 0.

Table 8. Mass balance calculations for ^{60}Co after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	2.13×10^6	7.45×10^4	54.2	2.5
	filter	2.59×10^4	5.57×10^3	0.8	0.2
b, inside control sac	filtrate	2.84×10^5	1.01×10^4	8.4	0.3
	filter	1.87×10^3	2.78×10^2	0.0	0.0
(2) Dialysis sac contents					
a, sediment sac	filtrate	4.71×10^4	1.56×10^3	1.4	0.1
	filter	1.20×10^6	6.59×10^3	30.5	0.7
b, plankton sac	filtrate	5.80×10^4	1.71×10^3	1.7	0.1
	filter	8.69×10^3	4.82×10^2	0.3	0.0
(3) Subsamples		1.76×10^5	7.18×10^2	4.5	0.1
(4) Flask washdown and dialysis sacs		5.45×10^1	8.2×10^{-1}	0.0	0.0
Total of (1), (2), (3), and (4)		3.93×10^6	7.57×10^4	101.8	2.7
Amount added+		3.62×10^6	9.13×10^4		
% Accounted for				108.6	3.5
% Unaccounted for				+ 8.6	3.5

*Single sample error values are one-sigma, propagated, counting error.

+Amount added = spiked water at day 0.

Table 9. Percent distribution of radionuclides that were recovered after 11 days of dialysis experiment.

Compartments		^{241}Am	^{137}Cs	^{238}Pu	^{207}Bi	^{65}Zn	^{60}Co
Spiked water	<0.22m	26.1	86.3	77.7	64.4	44.7	54.2
	>0.22m	8.2	0.0	3.0	0.0	0.8	0.8
Control Sac	<0.22m	0.7	2.2	0.5	0.0	1.0	8.4
	>0.22m	0.5	0.1	0.1	0.6	0.1	0.1
Sediment Sac	<0.22m	0.0	2.1	0.2	0.0	1.1	1.4
	>0.22m	47.1	1.6	9.2	22.3	25.8	30.5
Plankton Sac	<0.22m	0.8	2.2	0.4	1.9	2.4	1.7
	>0.22m	12.4	0.3	2.7	11.5	16.9	0.3
Samples during experiment		4.5	0.7	2.5	2.8	7.3	4.5
Miscellaneous apparatus		0.5	0.0	3.8	0.6	0.0	0.0

The final experiment to be discussed in this report was conducted to investigate the effect of organic detritus on the distribution of radionuclides and the rate of diffusion across the dialysis membranes. Filtered (<0.45 µm) water from the Skagit River estuary was spiked with ^{59}Fe, ^{60}Co, ^{106}Ru, ^{137}Cs, and ^{241}Am and allowed to equilibrate. Approximately 625 ml of this solution was placed in the outer compartment and four dialysis sacs containing 60 ml of filtered, but unspiked, water from the Skagit River estuary were submerged in the outer solution. The phytoplankton sac contained 1.2×10^7 cells of Phaeodactylum tricornutum, the sediment sac contained 12.8 mg of the pure clay montmorillonite, and the detritus sac contained 6.0 mg of dried and finely ground Zostera sp.; no additions were made to the control dialysis sac. The experiment was conducted at pH = 7.5, salinity = 32 °/oo and temperature = 20°C.

In order to obtain information on the initial rate of diffusion from the outer compartment into the dialysis sacs, samples were collected from each chamber at 2, 10, 24, 48, and 96 hours and the activity (cpm/ml) was determined for the soluble (<0.45 µm) and particulate (>0.45 µm) fractions. The distribution of radionuclides for each of the sampling times is given in Tables 10-14. By comparing the concentration of soluble radionuclides in each compartment we can estimate the time required to reach equilibrium between the outside chamber and the inside

Table 10. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co, and ^{59}Fe after two hours in a dialysis experiment

		Spiked Seawater	Dialysis Chambers			
			Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	152.5 (4.2)	5.56 (0.46)	1.36 (0.84)	2.02 (0.89)	1.08 (0.79)
	Particulate	705.3 (2.5	1.91 (0.47)	3.90 (0.53)	1.55 (0.23)	0.59 (0.45)
	% Particulate	82.2	25.6	74.1	43.4	35.3
^{137}Cs	Soluble	18.2 (0.92)	14.7 (0.40)	16.2 (0.76)	16.5 (0.76)	16.5 (0.72)
	Particulate	< 0.37	< 0.16	< 0.16	< 0.06	< 0.18
	% Particulate	< 2.0	< 1.1	< 1.0	< 0.4	< 1.1
^{106}Ru	Soluble	9.26 (0.76)	2.02 (0.21)	1.70 (0.41)	2.41 (0.45)	1.55 (0.43)
	Particulate	2.70 (0.37)	< 0.14	< 0.14	< 0.06	< 0.14
	% Particulate	22.6	< 6.5	< 7.6	< 2.3	< 8.3
^{65}Zn	Soluble	25.51 (0.97)	9.41 (0.32)	7.69 (0.55)	10.37 (0.62)	6.91 (0.52)
	Particulate	1.02 (0.30)	< 0.14	< 0.13	< 0.8	< 0.19
	% Particulate	3.8	< 1.15	< 1.7	< 7.2	< 2.7
^{60}Co	Soluble	45.98 (1.19)	16.32 (0.39)	8.79 (0.73)	17.09 (0.72)	17.11 (0.71)
	Particulate	2.61 (0.33)	< 0.12	< 0.13	< 0.06	< 0.14
	% Particulate	5.4	< 0.8	< 0.2	< 0.4	< 0.8
^{59}Fe	Soluble	11.01 (0.80)	0.31 (0.20)	0.81 (0.35)	0.44 (0.38)	0.45 (0.35)
	Particulate	30.31 (0.82)	< 0.10	< 0.11	< 0.05	0.15 (0.11)
	% Particulate	73.4	<24.4	<12.0	<10.2	25.0

Table 11. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after ten hours in a dialysis experiment.

			Dialysis Chambers			
		Spiked Seawater cpm/ml±2 S D	Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/ml±2 S D
^{241}Am	Soluble	116.5 (2.2)	15.79 (1.11)	18.19 (1.17)	15.55 (1.08)	18.10 (1.18)
	Particulate	451.5 (3.4)	11.47 (0.84)	49.20 (1.27)	21.81 (0.95)	43.94 (1.20)
	% Particulate	79.5	42.1	73.0	58.4	70.8
^{137}Cs	Soluble	21.5 (0.95)	20.3 (0.89)	21.6 (0.91)	20.9 (0.87)	19.2 (0.84)
	Particulate	< 0.30	< 0.19	< 0.20	< 0.19	< 0.19
	% Particulate	< 1.4	< 0.9	< 0.9	< 0.9	< 1.0
^{106}Ru	Soluble	5.45 (0.73)	4.65 (0.56)	4.31 (0.60)	4.24 (0.55)	4.01 (0.58)
	Particulate	1.86 (0.33)	0.17 (0.16)	1.46 (0.22)	< 0.18	0.35 (0.20)
	% Particulate	25.4	3.5	25.3	< 4.1	8.0
^{65}Zn	Soluble	23.65 (0.97)	17.67 (0.80)	5.63 (0.59)	13.66 (0.70)	7.28 (0.62)
	Particulate	0.33 (0.25)	0.21 (0.17)	0.41 (0.19)	0.51 (0.18)	1.45 (0.25)
	% Particulate	1.4	1.2	6.8	3.6	16.6
^{60}Co	Soluble	46.10 (1.20)	36.75 (1.07)	5.68 (1.10)	30.52 (0.97)	36.99 (0.97)
	Particulate	1.00 (0.28)	< 0.19	0.36 (0.21)	< 0.19	0.37 (0.21)
	% Particulate	2.1	< 0.5	6.0	< 0.6	1.0
^{65}Fe	Soluble	8.01 (0.74)	0.24 (0.13)	0.53 (0.25)	0.63 (0.47)	< 0.53
	Particulate	19.50 (0.66)	< 0.13	0.41 (0.14)	< 0.15	0.22 (0.16)
	% Particulate	70.9	<35.1	43.6	<19.2	> 29.3

Table 12. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 24 hours in a dialysis experiment.

		Spiked Seawater	Dialysis Chambers			
			Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	100.5 (2.0)	23.97 (1.26)	25.45 (1.31)	22.40 (1.24)	47.71 (1.55)
	Particulate	195.7 (2.3)	13.16 (0.68)	100.6 (1.7)	74.50 (1.50)	138.3 (1.9)
	% Particulate	66.1	35.4	79.8	76.9	74.4
^{137}Cs	Soluble	19.9 (0.93)	20.5 (0.90)	22.2 (0.88)	21.1 (0.93)	19.8 (0.93)
	Particulate	< 0.24	< 0.13	< 0.20	< 0.22	< 0.22
	% Particulate	< 1.4	< 0.6	< 0.9	< 1.1	< 1.1
^{106}Ru	Soluble	5.99 (0.66)	4.51 (0.63)	5.29 (0.60)	4.73 (0.62)	6.18 (0.62)
	Particulate	1.14 (0.25)	0.24 (0.12)	1.81 (0.25)	1.36 (0.21)	0.61 (0.28)
	% Particulate	16.0	5.0	25.5	22.3	9.0
^{65}Zn	Soluble	22.18 (0.92)	11.26 (0.71)	4.60 (0.59)	18.06 (0.85)	2.45 (0.57)
	Particulate	1.19 (0.23)	0.13 (0.12)	0.64 (0.19)	0.82 (0.23)	1.70 (0.24)
	% Particulate	5.1	1.1	12.2	4.3	41.0
^{60}Co	Soluble	41.89 (1.04)	43.63 (0.82)	4.76 (1.13)	36.68 (1.07)	44.28 (1.15)
	Particulate	0.46 (0.22)	< 0.13	0.48 (0.21)	< 0.22	0.37 (0.22)
	% Particulate	1.1	< 0.3	9.2	< 0.6	0.8
^{59}Fe	Soluble	6.76 (0.68)	1.22 (0.55)	1.06 (0.52)	2.20 (0.51)	1.55 (0.59)
	Particulate	8.26 (0.43)	0.44 (0.11)	1.13 (0.20)	1.20 (0.23)	0.86 (0.32)
	% Particulate	54.9	26.5	51.6	31.7	35.7

Table 13. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 48 hours in a dialysis experiment

			Dialysis Chambers			
		Spiked Seawater cpm/ml±2 S D	Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/ml±2 S D
^{241}Am	Soluble	101.7 (2.1)	27.73 (1.38)	41.60 (0.88)	26.20 (1.34)	110.5 (2.2)
	Particulate	108.2 (2.4)	39.24 (1.18)	219.1 (2.4)	98.80 (1.73)	272.5 (2.6)
	% Particulate	51.4	58.6	84.0	79.0	71.1
^{137}Cs	Soluble	20.2 (0.92)	18.9 (0.90)	20.9 (0.50)	21.8 (0.93)	21.9 (0.94)
	Particulate	<0.23	<0.19	<0.21	<0.20	<0.22
	% Particulate	<1.1	<1.0	<1.0	<0.9	<1.0
^{106}Ru	Soluble	6.15 (0.68)	5.55 (0.62)	5.10 (0.36)	6.29 (0.63)	7.07 (0.67)
	Particulate	0.48 (0.21)	0.22 (0.17)	3.40 (0.31)	1.11 (0.23)	0.82 (0.23)
	% Particulate	7.2	3.8	40.0	15.0	10.4
^{65}Zn	Soluble	23.54 (0.93)	16.48 (0.85)	14.84 (0.44)	22.98 (0.90)	11.13 (0.72)
	Particulate	0.58 (0.19)	0.29 (0.17)	0.87 (0.24)	1.17 (0.24)	1.76 (0.25)
	% Particulate	2.4	1.7	5.6	4.8	13.7
^{60}Co	Soluble	44.28 (1.18)	43.26 (1.14)	15.01 (0.65)	39.40 (1.10)	45.88 (1.19)
	Particulate	<0.23	<0.20	0.70 (0.20)	0.27 (0.22)	0.53 (0.24)
	% Particulate	<0.4	<0.5	4.5	0.7	1.14
^{54}Fe	Soluble	6.40 (0.67)	1.57 (0.59)	1.71 (0.31)	1.81 (0.56)	3.86 (0.66)
	Particulate	4.56 (0.34)	0.93 (0.18)	2.30 (0.28)	2.63 (0.28)	1.90 (0.26)
	% Particulate	41.6	37.2	57.4	59.2	33.0

Table 14. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 96 hours in a dialysis experiment.

			Dialysis Chambers			
		Spiked Seawater	Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	134.5 (2.3)	33.99 (0.85)	54.75 (1.71)	26.84 (1.40)	134.1 (2.4)
	Particulate	62.7 (1.4)	68.95 (1.40)	357.4 (3.0)	149.4 (1.1)	555.6 (3.7)
	% Particulate	31.8	66.3	86.7	84.8	80.6
^{137}Cs	Soluble	21.2 (0.92)	20.3 (0.51)	20.4 (0.95)	23.2 (0.93)	23.0 (0.93)
	Particulate	<0.19	<0.20	<0.23	<0.15	<0.24
	% Particulate	<0.9	<1.0	<1.1	<0.6	<1.1
^{106}Ru	Soluble	4.73 (0.68)	5.55 (0.35)	6.05 (0.65)	6.41 (0.62)	5.39 (0.68)
	Particulate	0.30 (0.18)	0.33 (0.20)	3.87 (0.34)	1.58 (0.16)	1.05 (0.26)
	% Particulate	6.0	5.6	39.0	19.8	16.3
^{65}Zn	Soluble	23.11 (0.93)	20.25 (0.48)	16.03 (0.82)	19.48 (0.85)	8.40 (0.69)
	Particulate	<0.19	0.47 (0.47)	1.50 (0.26)	1.87 (0.16)	1.94 (0.27)
	% Particulate	<0.8	2.3	8.6	8.8	18.8
^{60}Co	Soluble	42.04 (1.16)	47.13 (0.69)	16.21 (1.19)	37.76 (1.07)	47.40 (1.12)
	Particulate	<0.21	0.40 (0.22)	1.32 (0.20)	1.03 (0.16)	0.40 (0.23)
	% Particulate	<0.3	0.8	7.53	2.7	0.8
^{59}Fe	Soluble	5.44 (0.66)	1.73 (0.34)	2.49 (0.58)	1.93 (0.58)	3.11 (0.66)
	Particulate	2.96 (0.28)	1.95 (0.23)	5.43 (0.38)	6.53 (0.22)	5.38 (0.35)
	% Particulate	35.2	53.0	68.6	77.2	63.4

of the dialysis sacs for each radionuclide. Cesium-137 reaches equilibrium after 10 hours and ^{106}Ru reaches equilibrium after 24 hours. However for the radionuclides, ^{241}Am, ^{60}Co, ^{65}Zn, and ^{59}Fe, at least one dialysis sac does not come to equilibrium during the 96 hours of the experiment.

Radionuclides that occur primarily in ionic or other small molecular weight species are transported across the dialysis membranes very rapidly. For example, for ^{137}Cs (Fig. 9) all the dialysis sacs appear to be in equilibrium with the outer compartment after 10 hours and there are only small changes in concentration after that time. Table 15 shows that the concentration of particulate ^{137}Cs never exceeded the minimum detection limits and that less than 1% of the total ^{137}Cs could be in the particulate phase. Thus, for ^{137}Cs, transport across the dialysis membranes and equilibrium among all the experimental compartments is very rapid. The diffusion rate of other radionuclides into the dialysis sacs is dependent upon their physico-chemical state in the outer compartment. For example, ^{59}Fe and ^{241}Am which form a significant amount of particulates in the outer chamber do not reach equilibrium during this experiment.

For ^{241}Am there is a rapid decrease in the concentration in the outer compartment (Figs. 10 and 11) and an increase in both the soluble and particulate phases in all the dialysis sacs. However, the total concentration of ^{241}Am within the dialysis sacs continues to increase throughout the experiment and the soluble species do not come to equilibrium among the different compartments within the 96 hours of this experiment.

The highest concentrations of both particulate and soluble ^{241}Am are found in the detritus chamber followed by the phytoplankton chamber. This suggests that adsorption to suspended organic matter may be more important for the transport of ^{241}Am than sorption onto inorganic particulates. The affinity of ^{241}Am for organic surfaces indicates that chemical complexation with organic components on the particulates as well as adsorption may contribute to the uptake of ^{241}Am. Complexation with organic compounds may also affect the soluble concentrations in the phytoplankton and detritus chambers. Dissolved organic compounds, either from direct solubilization or excretion of exometabolites, complex with ^{241}Am and increase the soluble concentration. It is difficult to assess the potential effects of dissolved organics since there is very little information on the complexation of ^{241}Am by organic ligands (11).

Throughout the experiment the concentration of particulate ^{241}Am is greater than the soluble concentration except in the outside chamber after 96 hours. This occurs because ^{241}Am forms radiocolloids which are partly retained on the filters and because of the affinity of americium for surfaces. The decrease of particulate ^{241}Am in the outside chamber throughout the experiment results from dissolution of particles as the

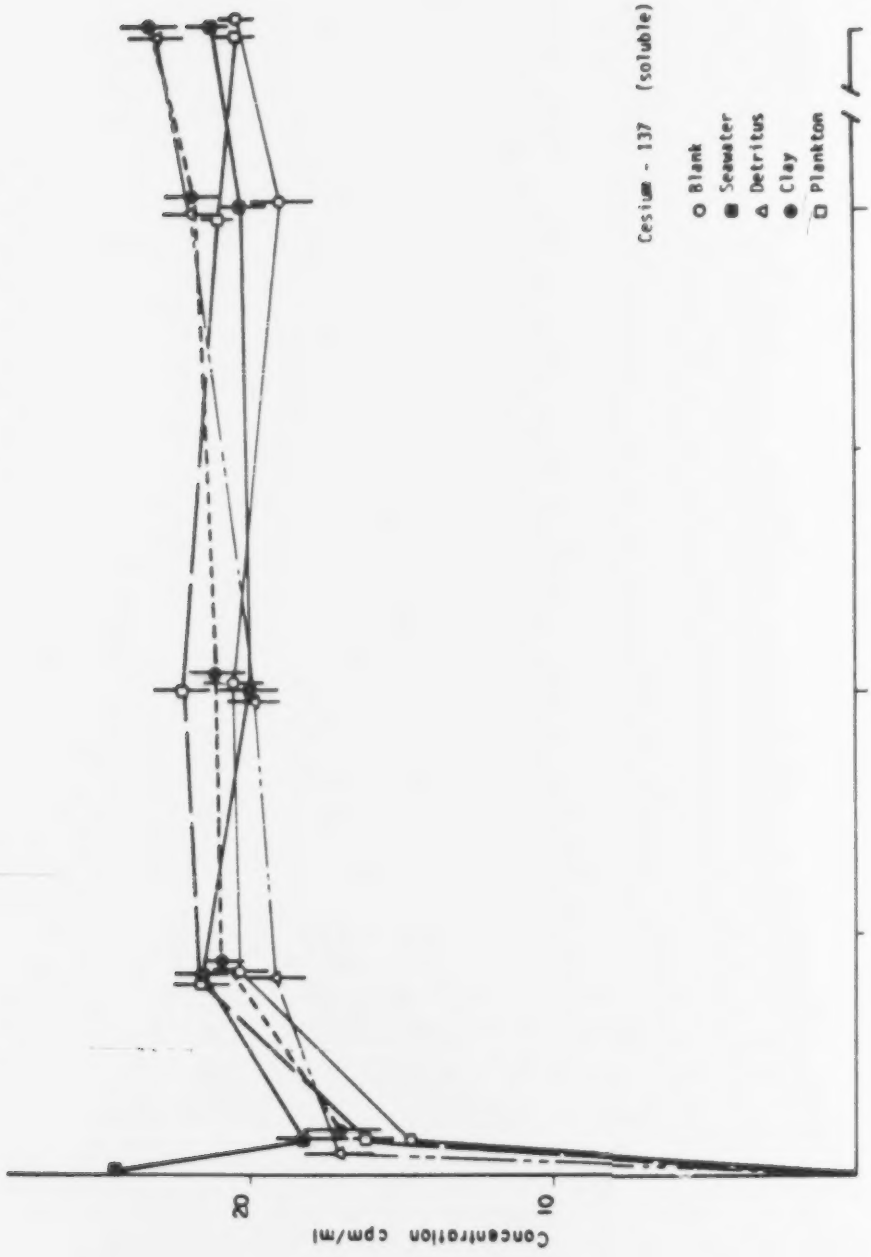

Figure 9 Concentration of soluble ^{137}Cs in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

Table 15. Distribution of ^{137}Cs during a 96 hour dialysis experiment.

Time (hrs.)		Spiked Seawater cpm/ml±2S.D.	Dialysis Chambers			
			Blank cpm/ml±2S.D.	Plankton cpm/ml±2S.D.	Clay cpm/ml±2S.D.	Detritus cpm/ml±2S.D.
0		24.4				
2	Soluble	17.7 (0.92)	14.2 (0.40)	15.7 (0.76)	16.0 (0.76)	16.0 (0.72)
	Particulate	0.36 (0.37)	0.47 (0.15)	0.30 (0.15)	0.35 (0.06)	0.50 (0.17)
	% Particulate	2.0	3.2	1.9	2.1	3.0
10	Soluble	20.9 (0.95)	19.7 (0.89)	21.0 (0.91)	20.3 (0.87)	18.6 (0.84)
	Particulate	0.52 (0.29)	0.20 (0.17)	0.60 (0.18)	0.22 (0.17)	0.73 (0.17)
	% Particulate	2.4	1.0	2.8	1.1	3.8
24	Soluble	19.3 (0.93)	19.9 (0.90)	21.6 (0.88)	20.5 (0.93)	19.2 (0.93)
	Particulate	0.51 (0.23)	0.40 (0.11)	0.60 (0.18)	0.49 (0.21)	0.30 (0.21)
	% Particulate	2.6	2.0	2.7	2.3	1.5
48	Soluble	19.6 (0.92)	18.3 (0.90)	20.3 (0.50)	21.2 (0.93)	21.3 (0.94)
	Particulate	0.45 (0.22)	0.49 (0.17)	0.73 (0.20)	0.75 (0.19)	0.59 (0.21)
	% Particulate	2.2	2.6	3.5	3.4	2.7
96	Soluble	20.6 (0.92)	19.7 (0.51)	19.8 (0.95)	22.6 (0.93)	22.4 (0.93)
	Particulate	0.27 (0.18)	0.72 (0.19)	0.56 (0.22)	0.60 (0.13)	0.55 (0.23)
	% Particulate	1.3	3.5	2.8	2.6	2.4

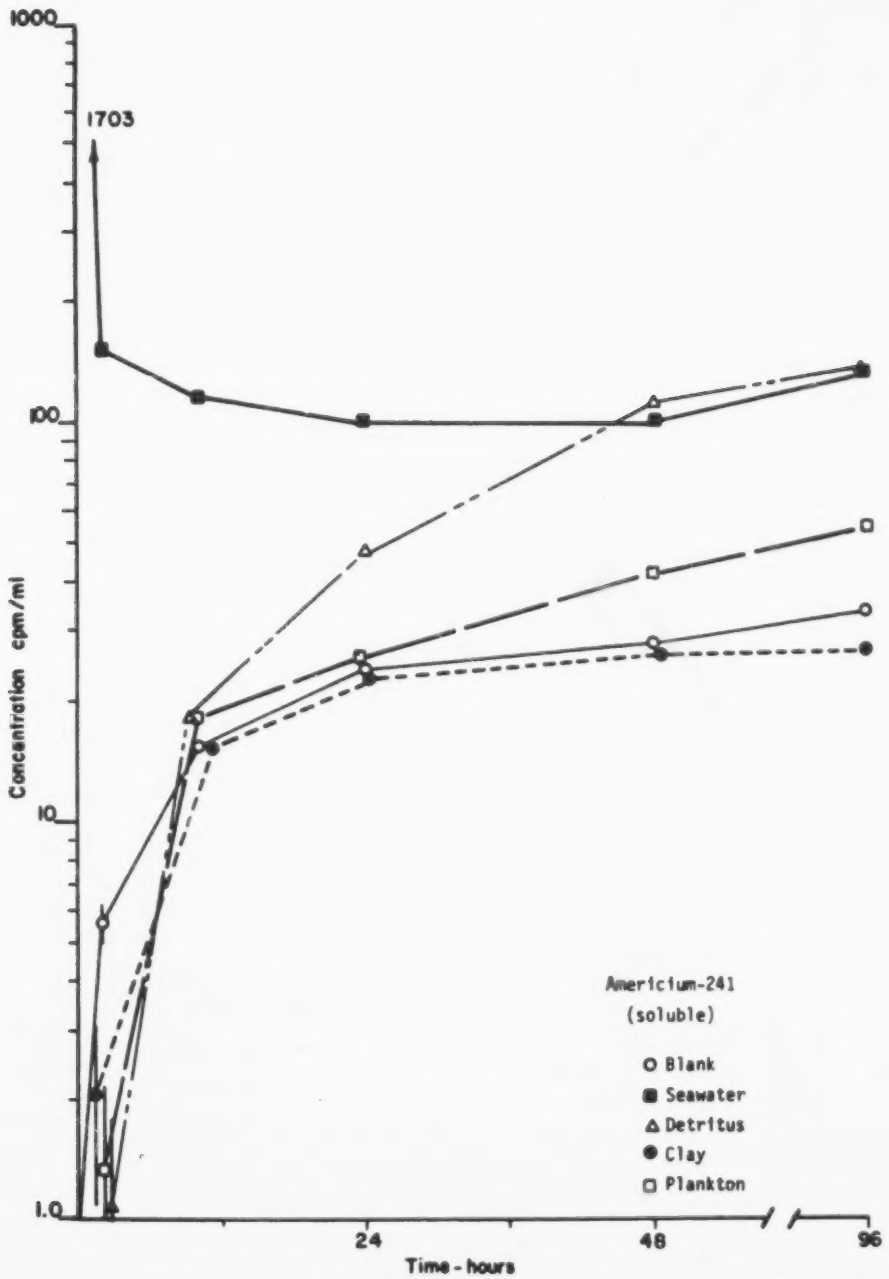

Figure 10. Concentration of soluble ^{241}Am in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

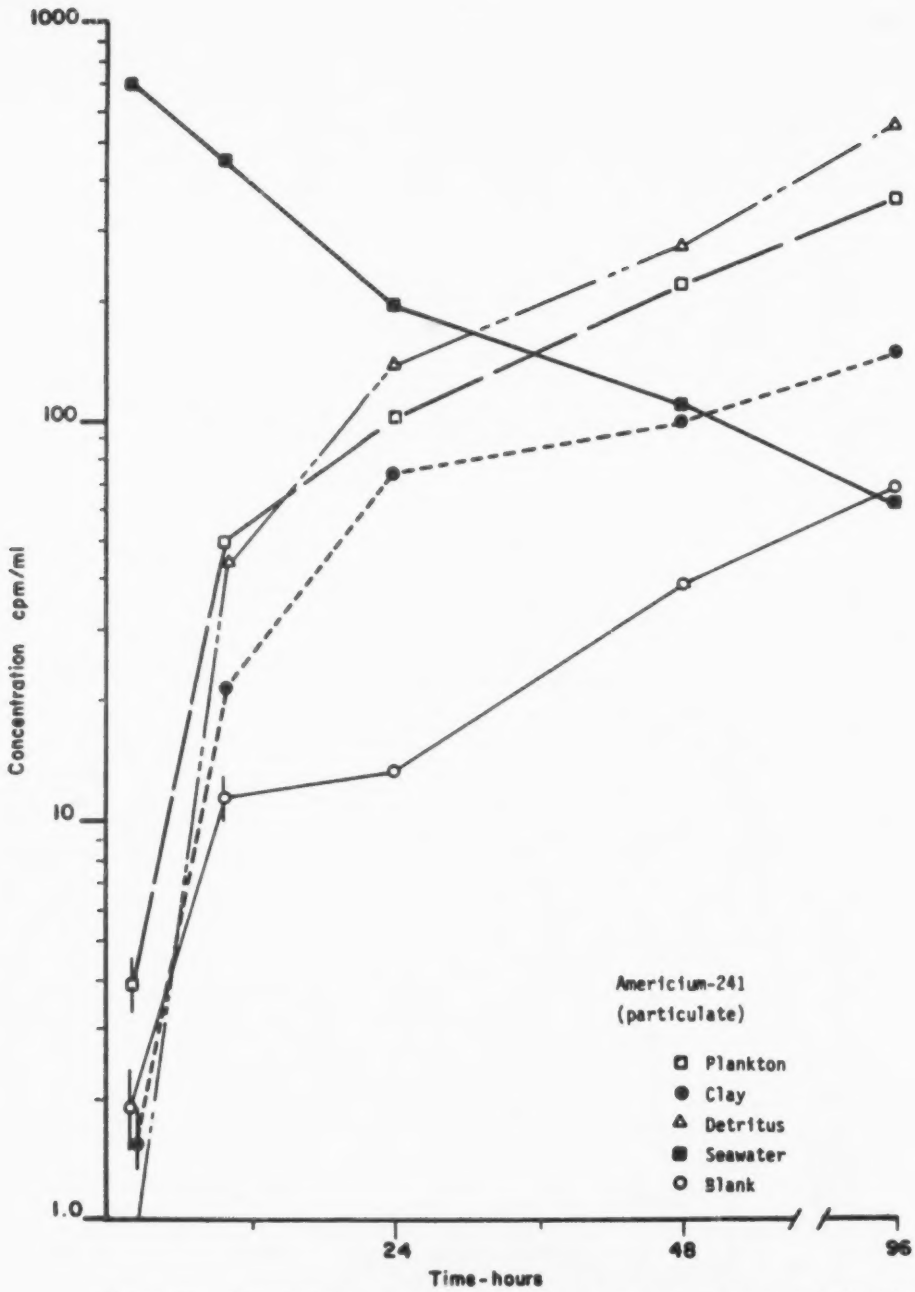

Figure 11. Concentration of particulate ^{241}Am in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

total concentration decreases or from sorption of particles to the beaker.

The data for ^{106}Ru are plotted in Figs. 12 and 13. Throughout the experiment there are higher concentrations of dissolved species than particulate species in all chambers. The concentration of ^{106}Ru in the particulate phase is generally less than 20% of the total concentration except for the dialysis sac containing plankton where approximately 40% of ^{106}Ru occurs in the particulate phase. The concentration of soluble ^{106}Ru species is comparable for each of the dialysis sacs after the first ten hours, but equilibrium with the outside chamber was not attained during that period. After 24 hours soluble ^{106}Ru in the detritus and plankton sacs appears to be in equilibrium with the outer compartment and the control and clay sacs reach equilibrium after 48 hours. Between 24 hours and 96 hours the dialysis sac with detritus and the outer spiked seawater compartment behave similarly, but the concentration of soluble species in the plankton, clay, and blank dialysis chambers change independently of the outer compartment. In particular the concentration of soluble ^{106}Ru in the detritus compartment and the outer chamber decrease between 48 hours and 96 hours while the concentrations in the other compartments either increase or remain unchanged. Differences among the compartments cannot be explained until we have better information about the chemical species that are present in each of the chambers. A possible explanation for the observed differences is that in some compartments soluble species may be formed which are not dialyzable.

The concentration of particulate ^{106}Ru (Fig. 13) in the outer compartment decreases throughout the experiment. This corresponds to the decrease observed for particulate ^{241}Am and suggests that both ^{241}Am and ^{106}Ru occur as radiocolloids or are adsorbed on colloids, such as iron oxides. We have shown previously (2) that in marine sediment-water systems significant amounts of ^{106}Ru and ^{241}Am occur in the particulate phase even if no suspended particles are added to the system. It seems likely that during the experiment these colloids adsorb to the dialysis membranes or to the glassware and the apparent concentration of particulate radionuclides, sorbed to the colloids, decrease. For both ^{106}Ru and ^{241}Am the concentration of particulates in the control dialysis sac and the outer chamber are equivalent after 96 hours again suggesting that the concentration of particulates for these two radionuclides is controlled by some other chemical species. Particulate ^{106}Ru does not appear within the dialysis sacs as quickly as the particulate forms of most other radionuclides. There was no significant concentration of particulate ^{106}Ru in any of the dialysis sacs after 2 hours and detectable concentrations in the clay chamber were not apparent until 24 hours. The clay, detritus and plankton chambers all showed significant increases in the concentration of ^{106}Ru in particulate phases during the experiment and had significantly higher concentrations of particulate ^{106}Ru than either the blank dialysis sac or the outer compartment.

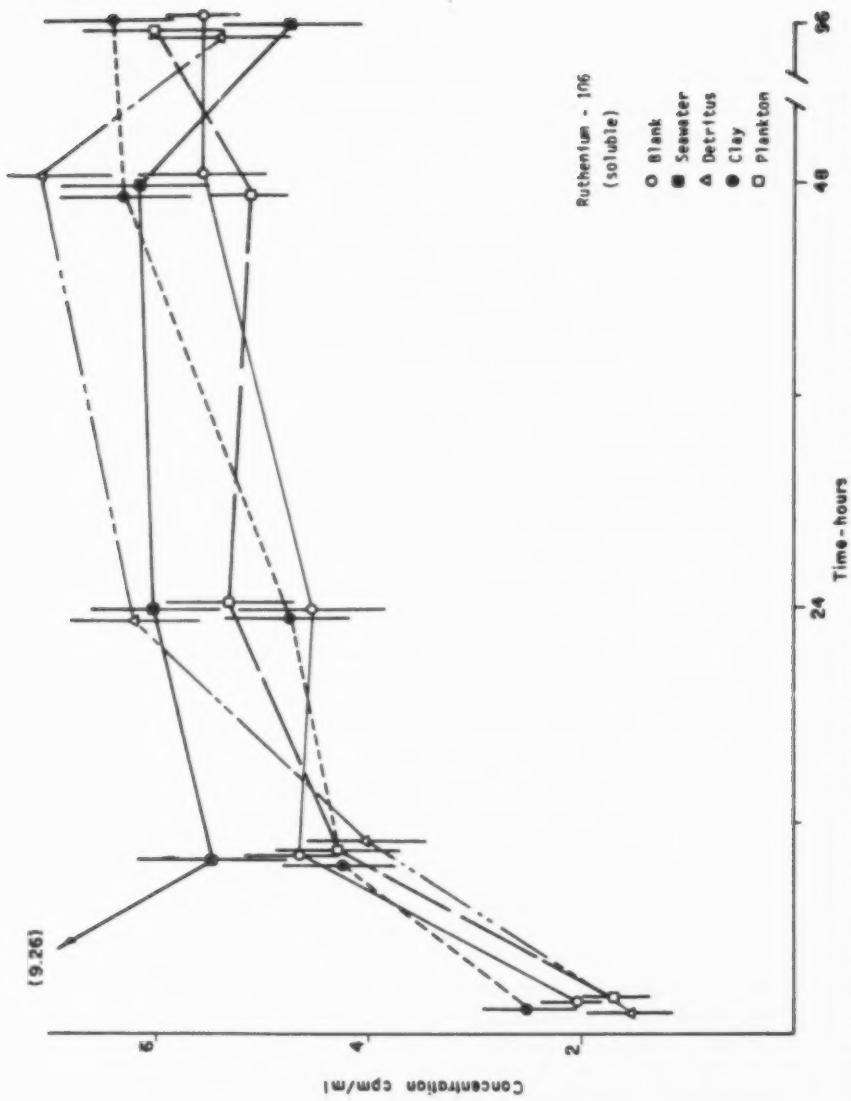

Figure 12. Concentration of soluble ^{106}Ru in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

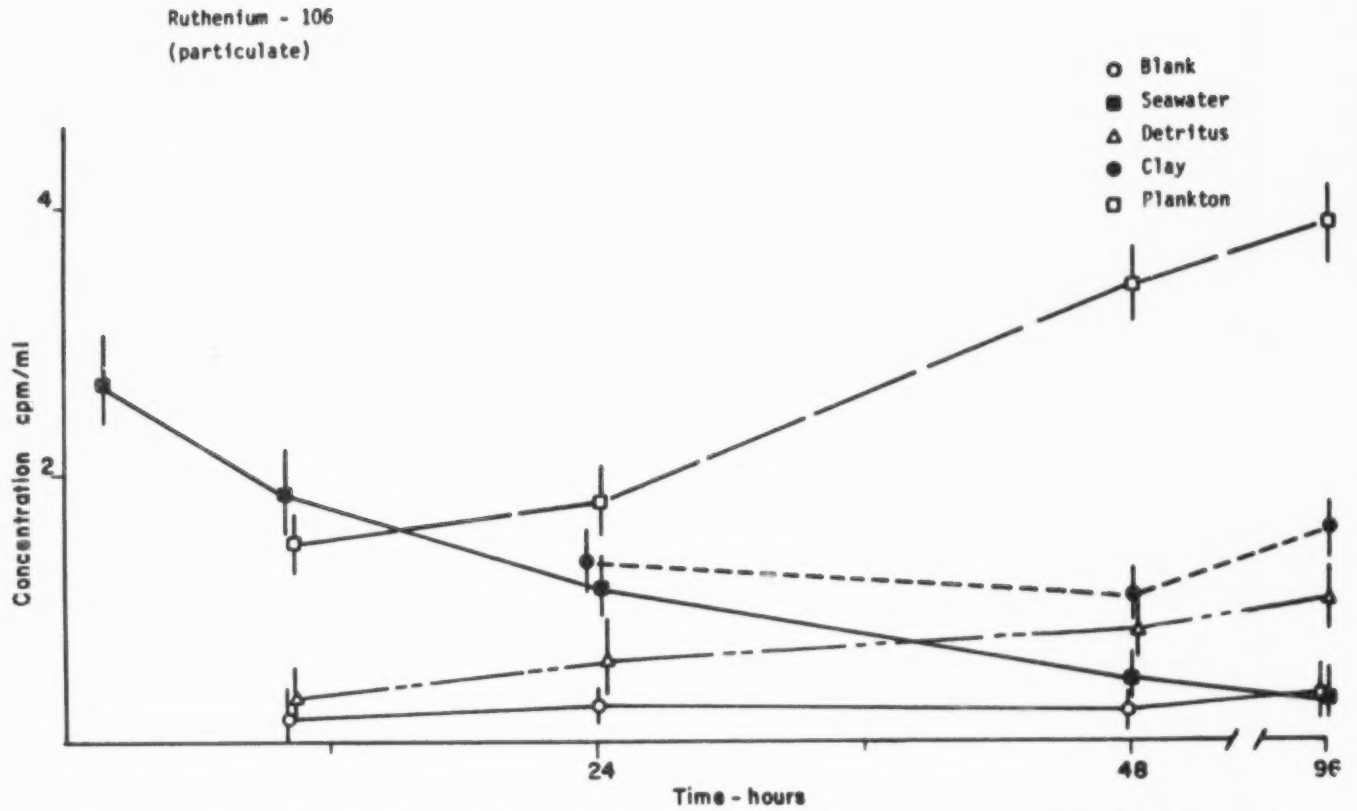

Figure 13. Concentration of particulate ^{106}Ru in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

Thus, each of the suspended solids take up ^{106}Ru. Phytoplankton showed the greatest affinity for ^{106}Ru followed by clay and detritus.

Of the radionuclides included in this experiment the data for ^{65}Zn are most variable. Figure 14 indicates that for soluble ^{65}Zn only the clay chamber reaches equilibrium with the outer chamber during the course of the experiment although the blank dialysis sac is approaching equilibrium 96 hours. This relatively slow approach to equilibrium is surprising because zinc occurs primarily in the dissolved phase, and is expected to occur predominantly as small inorganic species (12,13).

It has been found previously that the reaction rates for different physico-chemical species of zinc are on the order of days (14) and that ^{65}Zn does not exchange rapidly with the stable zinc present in seawater (15). It may be, therefore, that the results obtained in these experiments are not indicative of the behavior of ^{65}Zn over longer time periods in natural environments.

Throughout the experiment there are significant differences in the concentration of soluble ^{65}Zn species among the different dialysis sacs. In the first two hours; however, there is quite rapid exchange of ^{65}Zn between the outer compartment and each of the dialysis sacs indicating a rapid diffusion across the dialysis membranes for some chemical species. Therefore, it is unlikely that differential diffusion from the outer compartment into the dialysis sacs contributes significantly to the observed differences in the concentration of soluble ^{65}Zn. Rather, the concentration within each dialysis sac must be somewhat independent of the other chambers due to differences in chemical speciation among the different compartments.

Soluble ^{65}Zn concentration in the plankton and detritus chambers behave similarly throughout the experiment suggesting that organic ligands in these compartments may be contributing to the slow approach to equilibrium that is observed among the various chambers. Unfortunately to explain the lower concentrations of soluble ^{65}Zn in the plankton and detritus chambers relative to the other compartments we need to assume the formation of an organic complex which readily diffuses out of the chamber where it is formed but does not enter the other dialysis sacs. It is difficult to envision a mechanism that would behave in that manner. Therefore, the best explanation for the results obtained in this experiment is that isotopic disequilibrium between stable zinc and ^{65}Zn persists throughout the experiment. This could occur if stable zinc was introduced into the experiment with the plankton and detritus and solubilized during the experiment. Then, the total zinc concentration inside those dialysis sacs would be higher and diffusion of ^{65}Zn would be retarded.

The particulate species of ^{65}Zn (Fig. 15) behave more predictably than the soluble species ^{65}Zn. All of the dialysis chambers have detectable concentrations of particulate ^{65}Zn after ten hours, and, except for

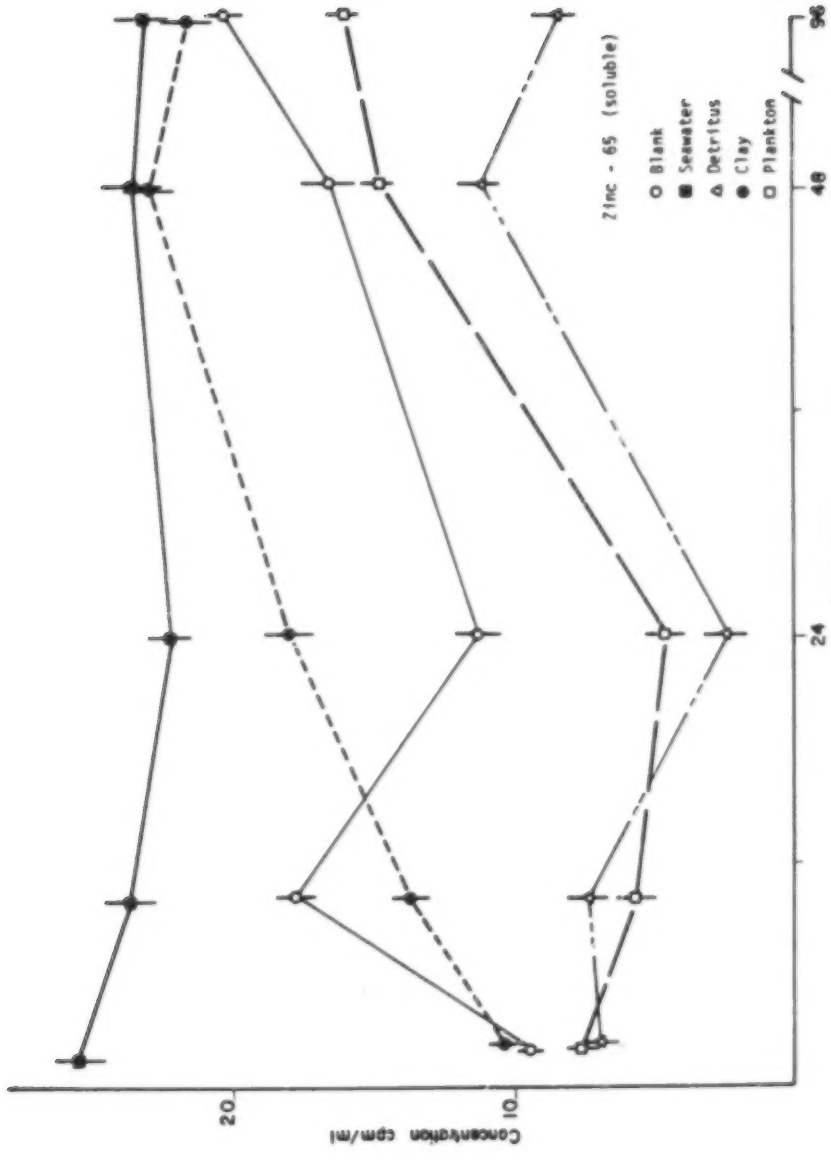

Figure 14. Concentration of soluble ^{65}Zn in each compartment at 2, 10, 24, 43 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

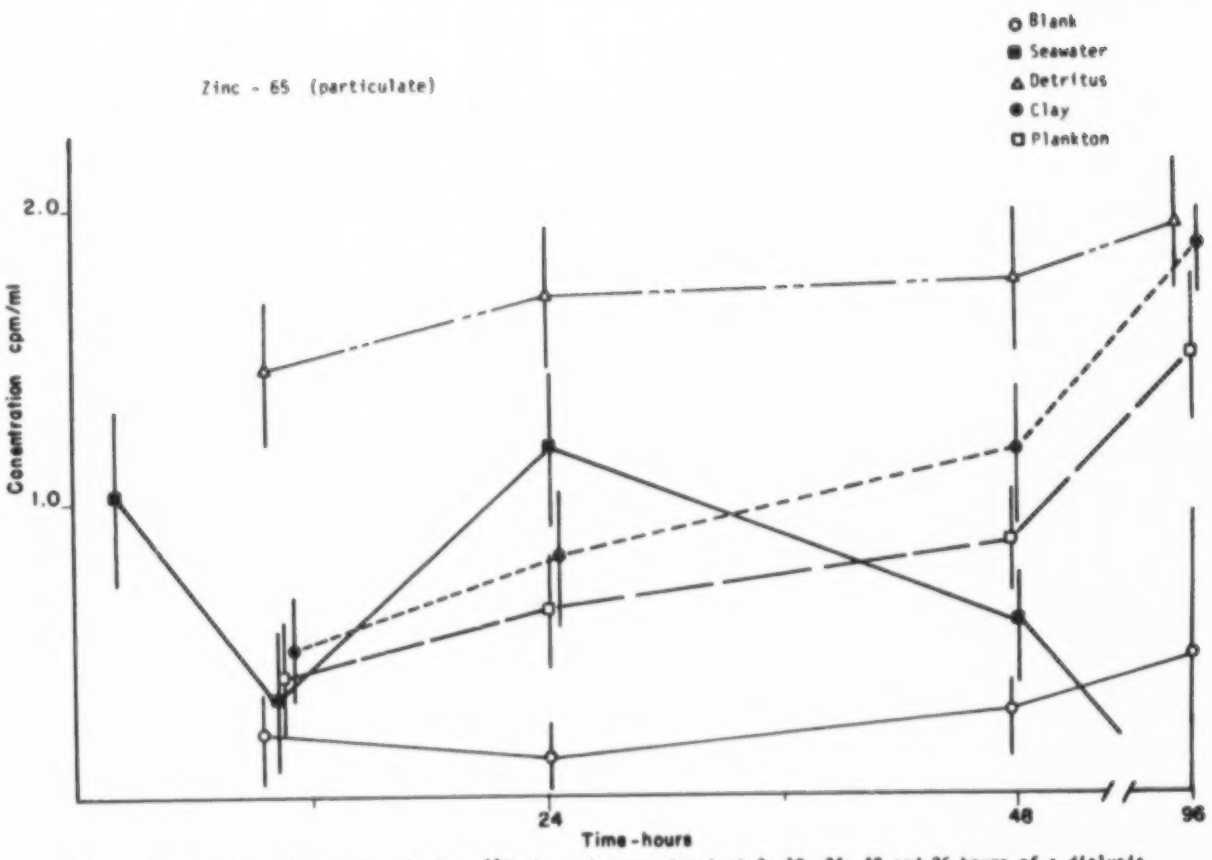

Figure 15. Concentration of particulate ^{65}Zn in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

the blank chamber, the concentration of particulate species increases throughout the experiment. Initially, after 10 hours, the concentration of particulate ^{65}Zn in the detrital chamber is significantly higher than for the clay or plankton sacs. However, after 96 hours all three chambers with suspended particulates have comparable concentrations. Furthermore, after 96 hours the concentration of particulate ^{65}Zn in those chambers is significantly higher than the concentration in the control dialysis sac or in the outside chamber. Therefore, a measurable amount of adsorption occurs on all three of the introduced particulates but there is no significant difference in the amount adsorbed (per milliliter) for any of the particulates. The concentration of particulate ^{65}Zn in the control dialysis sac is unchanged during the experiment and may in fact be an artifact resulting from the sorption of soluble species onto the membrane filter. It is, in any case, only a small portion of the total ^{65}Zn that diffused into the control dialysis sac. The concentration of particulate ^{65}Zn in the outer compartment decreases between 24 hours and 96 hours similarly to ^{106}Ru and ^{241}Am. In all compartments, the concentration of particulate ^{65}Zn is quite small relative to the soluble concentration.

While it is clear that zinc sorbs to the suspended particulates that were introduced into the experiment, soluble species of ^{65}Zn predominate in all chambers thoughout the experiment. These experiments indicate that particulate ^{65}Zn may be insignificant geochemically. However, adsorption of ^{65}Zn to organic particulates may introduce the radionuclide into marine food chains.

The concentrations of soluble ^{60}Co in each of the experimental compartments is plotted in Fig. 16. Although diffusion of ^{60}Co is not as rapid as for ^{137}Cs there are detectable concentrations in all four dialysis sacs after two hours. Equilibrium among the various compartments is reached within 24 hours except for the clay chamber which has a slightly lower concentration throughout the experiment. After 96 hours the concentration of soluble ^{60}Co in the clay chamber and the outside compartment appear to be converging at a somewhat lower concentration than in the other chambers. It is not known if this represents a significant difference or merely variation in the sampling.

Clay, plankton, and detritus particles all take up ^{60}Co (Fig. 17) but the concentration in the particulate phase is generally less than 1% of the total concentration. As with the other radionuclides the concentration of particulate ^{60}Co in the outer compartment decreases steadily throughout the experiment.

The behavior of ^{59}Fe is different than any other radionuclide that was included in the experiments. The concentration of soluble ^{59}Fe in the dialysis sacs does not approach the concentration in the outer compartment during the experiment (Fig. 18). However, the concentrations among the dialysis sacs are equivalent except for the value in the detritus sac after 48 hours. Since the values within the dialysis sacs are

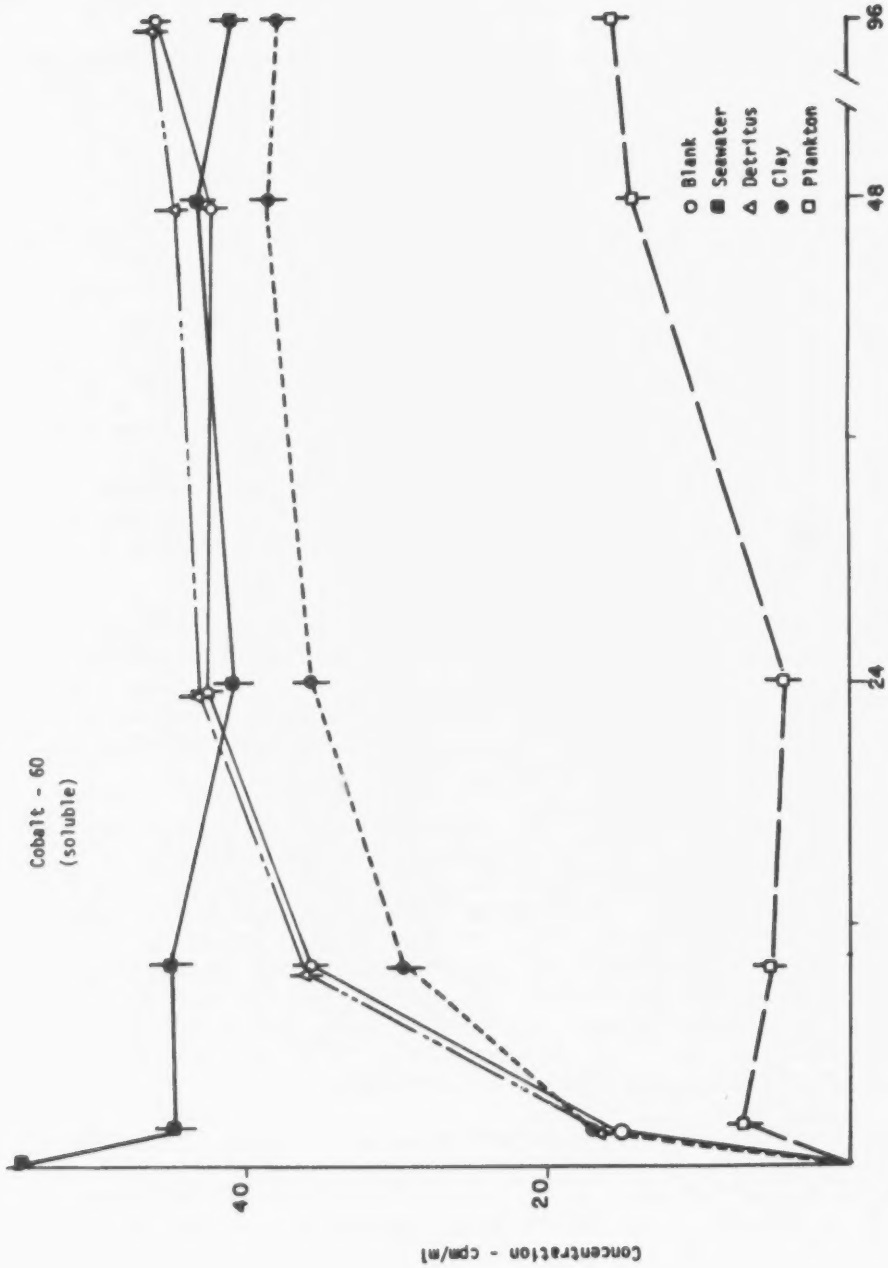

Figure 16. Concentration of soluble ^{60}Co in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

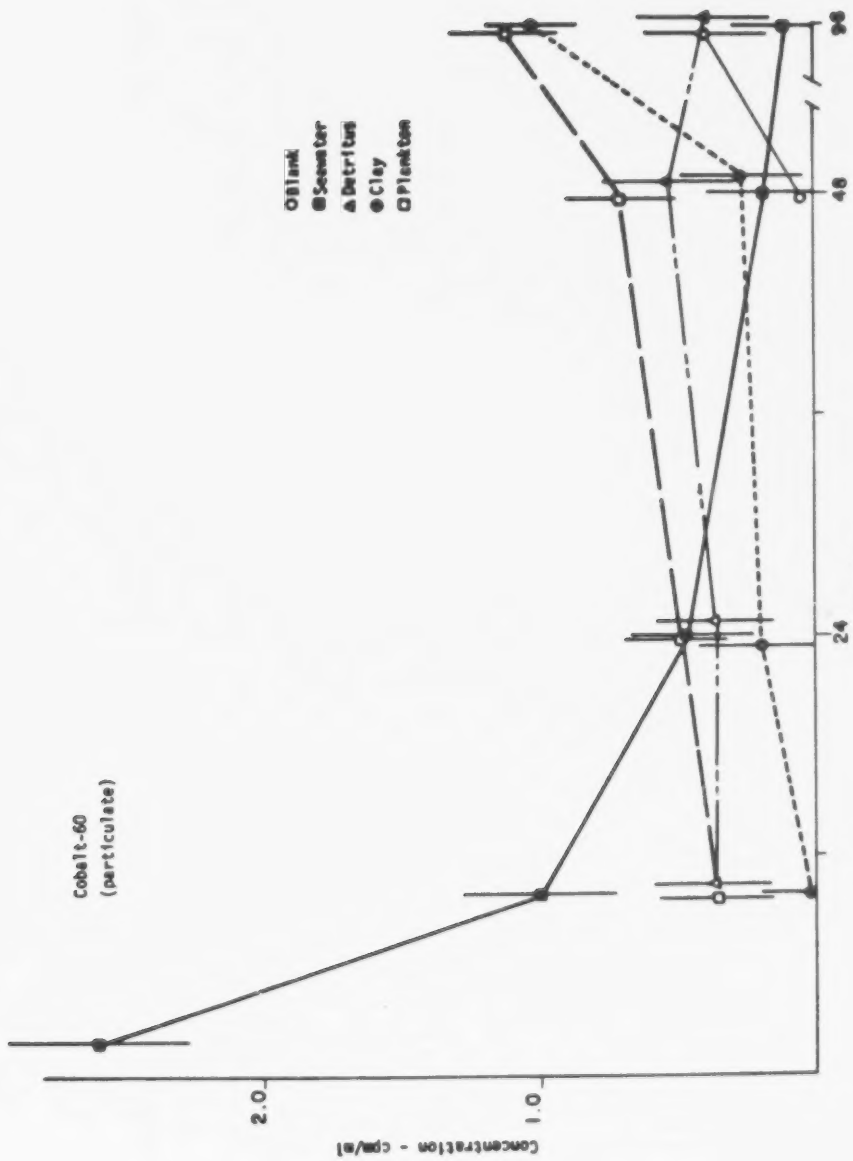

Figure 17. Concentrations of particulate ^{60}Co in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

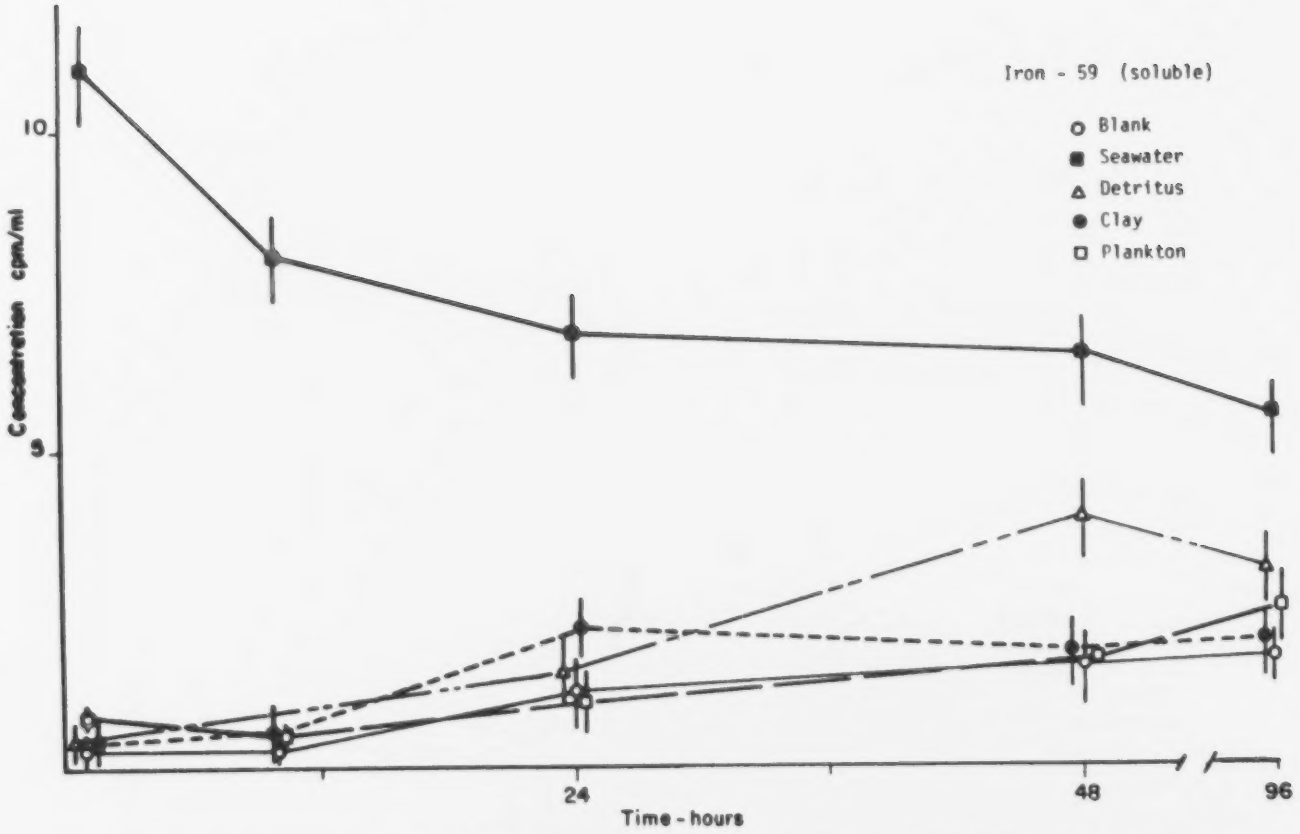

Figure 18. Concentrations of soluble ^{59}Fe in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

so consistent it suggests that truly soluble iron species are diffusing easily across the dialysis membrane and probably are in equilibrium throughout the experimental system. The higher concentrations that are reported for the outside chamber are most likely the result of colloidal iron species (16) that pass through the 0.45 μm Millipore filters but are too large to diffuse through the dialysis membranes. We have known for many years (17) that the concentration of filterable iron in seawater exceeds the solubility of expected inorganic solid precipitates. The difference between the filterable concentration and the predicted concentration of soluble iron was ascribed to the presence of colloidal iron. In these experiments the use of ^{59}Fe as an iron tracer allows us to observe the differences between filterable and dialyzable concentrations; they differ by at least a factor of two and perhaps as much as an order of magnitude.

The concentration of particulate ^{59}Fe in the outside compartment (Fig. 19) is initially much higher than the soluble concentration but declines steadily during the experiment. The decrease in particulate ^{59}Fe concentration probably occurs because the particles adsorb to the beaker and on the dialysis membranes. Although we did not calculate a mass balance budget for this experiment we did analyze the dialysis membranes and acid washes of the beaker after the experiment was completed. Those results indicate that in excess of 75% of the iron adsorbs to surfaces of the experimental apparatus.

The decrease in particulate ^{59}Fe is very similar to the decrease observed for the other radionuclides discussed previously. We believe the similar behavior for the particulates of all radionuclides in the outer compartment occurs because iron oxide solids precipitate out of solution and soluble species of the other radionuclides adsorb to the solid precipitates. As the iron solids adsorb to surfaces of the experimental apparatus the concentration of particulate ^{59}Fe decreases and the particulate concentration of other radionuclides that were adsorbed to the iron precipitates decreases concurrently. The radionuclides of interest in this experiment, ^{137}Cs, ^{241}Am, ^{106}Ru, and ^{60}Co all behave similarly in the outside compartment and the radionuclide concentrations in the particulate phase appear to be controlled by the presence of iron oxide solids.

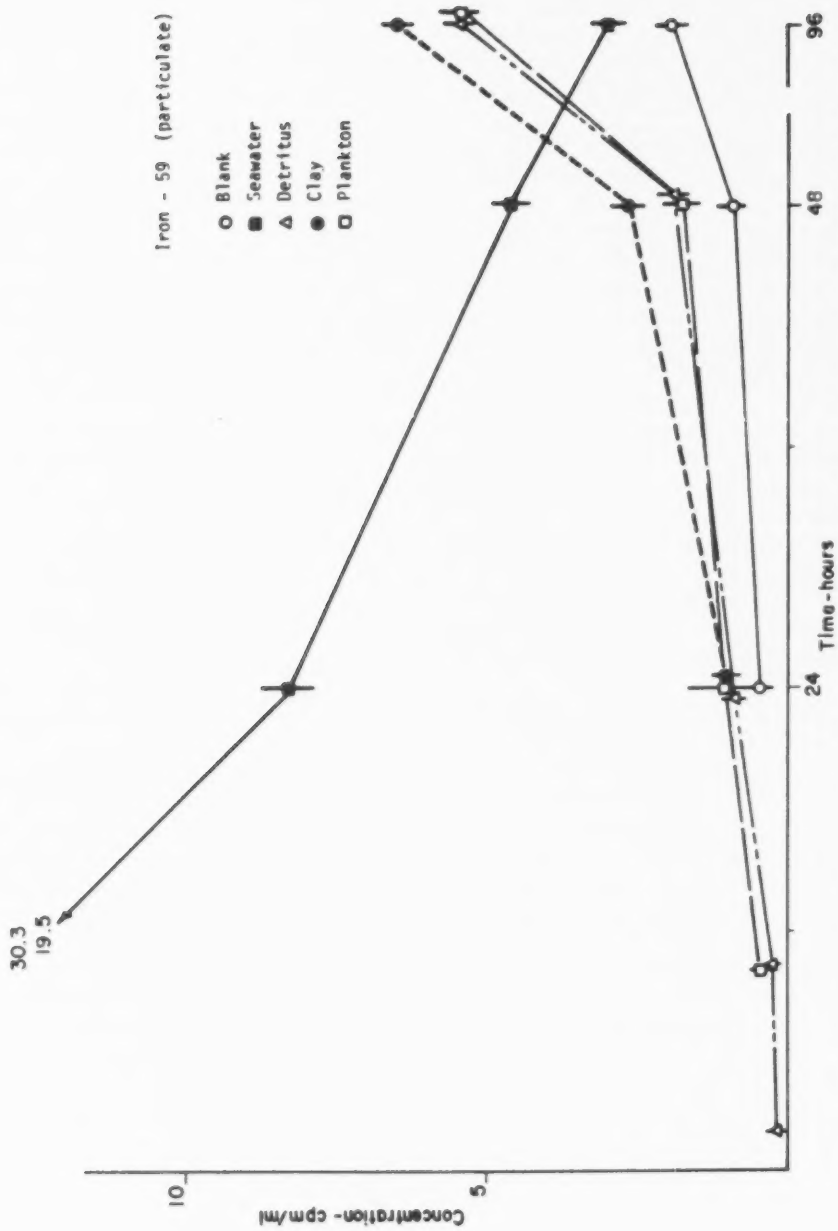

Figure 19. Concentration of particulate ^{59}Fe in each compartment at 2, 10, 24, 43 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

CONCLUSIONS AND RECOMMENDATIONS

1. Dialysis experiments present a valuable experimental approach for comparing the accumulation of radionuclides on different types of suspended particulates.

2. In these experiments most radionuclides are transported across the 6000-8000 molecular weight exclusion size dialysis membranes quite rapidly. For example, ^{137}Cs comes to equilibrium among the dialysis sacs and the outer compartment within 10 hours and ^{106}Ru reaches equilibrium during the first day. For these radionuclides it is easy to assess the relative accumulation on different types of suspended particulates.

3. Those radionuclides that occur primarily as particulates in the filtered seawater such as ^{59}Fe and ^{207}Bi diffuse slowly across the dialysis membranes. Since the soluble species of these radionuclides that pass across the dialysis membrane may precipitate inside the dialysis sac it is difficult to discriminate between the accumulation of these soluble radionuclides on the suspended particulates and the precipitation of particulate phases.

4. During a four day dialysis experiment ^{59}Fe, ^{60}Co, ^{65}Zn, ^{207}Bi, and ^{241}Am did not reach equilibrium between the outer compartment and the dialysis sacs. It appears that interactions among the different chambers, especially the formation of organic complexes in some compartments may contribute to the slow approach to equilibrium.

5. These experiments were conducted with dialysis membranes having a 6000-8000 molecular weight exclusion size. Therefore, we were able to evaluate the adsorption of soluble (<8000 mol. wt.) chemical species. Experiments with dialysis membranes having different nominal pore sizes should be conducted to obtain further information on the size distribution of the soluble species and the adsorption behavior of different size classes of physico-chemical species.

6. All of the radionuclides accumulate on the suspended particulates although the distribution depends upon the specific type and concentration of particulates that are introduced into an experiment. Additional experiments with pure clays, different types of detrital particles and other species of phytoplankton will provide more information on the relative affinity of radionuclides for various types of suspended particulates.

7. The affinity of some radionuclides, e.g., ^{106}Ru and ^{241}Am, for phytoplankton and detritus suggests that organic particulates may be an important factor in determining the eventual fate of these radionuclides. In addition, organic particles may provide a mechanisms for introducing radionuclides into aquatic food chains.

8. The adsorption behavior of ^{241}Am and ^{65}Zn appears to be affected by the presence of organic ligands in solution.

9. Additional research should be conducted to quantify the interactions of both dissolved and particulate organic matter with aquatic radionuclides and to evaluate the potential effects of organic matter in determining the eventual fate of introduced radionuclides.

10. Mass balance determinations show that we can account for nearly all of the ^{60}Co, ^{65}Zn, ^{137}Cs, and ^{207}Bi. However, ^{241}Am and ^{238}Pu which adsorb strongly to the experimental apparatus were not completely recovered.

11. Additional research should be conducted to determine the actual size distribution of colloidal and particulate radionuclides.

12. Additional research should be conducted to determine the effect of solid precipitates such as iron oxides or manganese dioxides on the partitioning of radionuclides between soluble and particulate phases.

SUMMARY

Radionuclides in natural aquatic environments may occur in a variety of different physico-chemical states. A convenient operational distinction has been made between dissolved (less than 0.45 μm) and particulate (greater than 0.45 μm) species. For many metals, including the radionuclides of interest, high concentrations are found in the particulate phase. These concentrations may result from adsorption of metals onto suspended particulates or colloidal particles or from solid precipitation. The principal objective of this research was to evaluate the distribution of selected radionuclides, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{106}Ru, ^{137}Cs, ^{207}Bi, ^{238}Pu, and ^{241}Am, between the particulate and soluble phases in marine environments and to investigate the affinity of radionuclides for different types of suspended particulate matter. Three different experiments were conducted to determine the adsorption kinetics and equilibrium distribution of radionuclides on suspended sediments, phytoplankton, and organic detritus. Radionuclides were equilibrated with filtered seawater for at least 10 days. Particulates were suspended in filtered seawater inside dialysis membranes. (6000-8000 molecular weight exclusion size) which were then introduced into the labeled seawater. Most radionuclides diffuse rapidly, within 24 hours, across the dialysis membrane and adsorb on the suspended particulates, although ^{59}Fe and ^{207}Bi do not reach equilibrium within the first four days. All radionuclides adsorb to some extent on the suspended particulates although ^{137}Cs occurs primarily in the soluble phase in all experiments. For ^{60}Co, ^{65}Zn, and ^{106}Ru, the higher concentrations sometimes occur in the soluble phase and sometimes in the particulate while for ^{59}Fe, ^{207}Bi, ^{238}Pu, and ^{241}Am the particulate phase always dominates. The extent of adsorption depends upon the radionuclide and specific particulates present in the experiment. For example, most radionuclides adsorb more strongly to natural sediments from Lake Nitinat than to the pure clay, calcium montmorillonite. The affinity of ^{106}Ru and ^{241}Am for phytoplankton and detritus suggests that organic particulates may have a significant influence on the eventual fate of some radionuclides. Additional experiments should be conducted using dialysis membranes with different pore sizes to evaluate the adsorption behavior of different physico-chemical species of radionuclides and with other types of suspended particulates to evaluate the affinity of radionuclides for different types of solid surfaces.

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16. ABSTRACT (200 words or less)

The overall objective of this research program was to obtain new information that can be used to predict the fate of radionuclides that may enter the aquatic environment from nuclear power plants, waste storage facilities or fuel reprocessing plants. Important parameters for determining fate are the distribution of radionuclides between the soluble and particulate phases and the partitioning of radionuclides among various suspended particulates. This report presents the results of dialysis experiments that were used to study the distribution of radionuclides among suspended sediments, phytoplankton, organic detritus, and filtered sea water. Three experiments were conducted to investigate the adsorption kinetics and equilibrium distribution of ^{59}Fe, ^{60}Co, ^{65}Zn, ^{106}Ru, ^{137}Cs, ^{207}Bi, ^{238}Pu, and ^{241}Am in marine system. Diffusion across the dialysis membranes depends upon the physico-chemical form of the radionuclides, proceeding quite rapidly for ionic species of ^{137}Cs and ^{60}Co but much more slowly for radionuclides which occur primarily as colloids and solid precipitates such as ^{59}Fe, ^{207}Bi, and ^{241}Am. All the radionuclides adsorb to suspended particulates although the amount of adsorption depends upon the specific types and concentration of particulates in the system and the selected radionuclide. High affinity of some radionuclides--e.g., ^{106}Ru and ^{241}Am--for detritus and phytoplankton suggests that suspended organics may significantly affect the eventual fate of those radionuclides in marine ecosystems.

17. KEY WORDS AND DOCUMENT ANALYSIS
Distribution Coefficients
^{106}Ru, ^{137}Cs, ^{241}Am,
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